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THE EFFECTS OF CRYSTALLINE BINDING
ON THE DOPPLER BROADENING OF A NEUTRON RESONANCE

BROTHER AUSTIN BERNABEI, F.S.C.

MASTER

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THE EFFECTS OF CRYSTALLINE BINDING ON THE
DOPPLER BROADENING OF A NEUTRON RESONANCE

by

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A dissertation in the Department of Physics submitted to
the Faculty of the Graduate School of Arts and Sciences in
partial fulfillment of the requirements for the degree of
Doctor of Philosophy.

New York University

April 1, 1964

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TO MY MOTHER
AND
TO THE MEMORY OF MY FATHER
AND
TO REV. BROTHER AUGUSTINE PHILIP, F.S.C.

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ABSTRACT

The effects of crystalline binding on the Doppler broadening of the 4.14 ev neutron resonance in tungsten metal have been experimentally investigated. The shape of the resonance was measured in great detail at temperatures of 4.2°K, 77.8°K, 296°K and 825°K using a neutron crystal spectrometer having good resolution.

The method of "shape analysis" was used to fit the 825°K data to the Breit-Wigner one-level formula. The following resonance parameters were obtained using the effective temperature model of Lamb to account for the Doppler broadening effect: $E_0 = 4.1422 \pm 0.0005$ ev, $\Gamma_\gamma = 55.5 \pm 0.5$ mev, and $\Gamma_m = 1.50 \pm 0.01$ mev. The analysis, performed on a high speed computer, employed none of the approximations which have been normally used to simplify analysis. The errors quoted came from the analysis alone and do not include systematic uncertainties.

In the past most resonance analysis have employed either the ideal gas model or the Lamb effective temperature model to correct for Doppler broadening. Two additional theories have been presented recently: one theory is by Nelkin and Parks and the other is by Egelstaff and Holt. Both these theories have been compared with the experimental results of this dissertation.

As the temperature of the metal target was lowered asymmetrical distortions in the shape were observed as well as shifts in the "apparent" resonant energy, E_0 , to lower energies. Neither the effective temperature theory of Lamb nor the Nelkin-Parks theory predicted these shifts since they contain no parameters to account for the temperature variation of the recoil energy. The Egelstaff-Holt theory did demonstrate

"apparent" E_0 shifts, but their results over-predicted these energy changes.

Lamb's theory showed good but not perfect agreement with the observed resonance shape at 825°K. As the temperature was reduced the disagreement between theory and data became more pronounced. At all temperatures Lamb's effective temperature model yielded a resonance shape that was in first order agreement with the data.

The expansion equation of Nelkin and Parks was slightly superior to the Lamb theory for predicting the experimental resonance shape since the first term of the expansion is equivalent to Lamb's theory. The resonance shape derived from the Nelkin-Parks theory at 825°K gave an excellent fit to the observed data. The results indicated that future resonance studies directed to determining Breit-Wigner parameters to high accuracy should use this theory to correct for the Doppler broadening.

Preliminary comparisons of the data with the theory of Egelstaff and Holt were made. The calculations were carried out with computer programs LEAP AND LINESHAPE of R.C.F. McLatchie using a Debye frequency spectrum for the tungsten lattice vibrations. At temperatures of 825°K and 296°K the results of this theory are in very good agreement with the experimental data. Calculations based on this theory which employ an experimentally determined frequency spectrum have not yet been carried out but should be possible in the near future as soon as the data becomes available.

In addition to the metal target, a sample in the form of a heavy water solution of sodium tungstate was also measured at room temperature. Comparison of the observed line shapes for these two different

targets showed distinct differences which could not be explained by treating the solution as an ideal gas.

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CHAPTER I

INTRODUCTION

A. Effects of Thermal Motion on Resonances in Neutron Cross Sections:

The purpose of this dissertation is to present the results of an experimental study on the Doppler broadening of a neutron resonance. The observed data have been compared with several theories offered to explain this effect.

Neutron cross section measurements made on intermediate and heavy isotopes reveal that they contain many resonances. In general these resonances may be characterized as being narrow and closely spaced. In many instances their peak values are several thousand times greater than the geometrical cross section. The resonance spectra for different isotopes show wide variations in strength and spacing. For a given isotope the resonance structure is an intrinsic property by which the isotope may be identified.

Each resonance represents an excitation state of the compound nucleus formed after the addition of a neutron to a capturing nucleus. Each of these nuclear states has definite properties which can be determined by a careful study made of them. During the past two decades these levels have been studied extensively

both experimentally¹ and theoretically². The theoretical shape of an isolated resonance is expressed mathematically by the Breit-Wigner³ single-level formula.

The measured shape of a resonance differs from the natural shape because of the effects of Doppler broadening. The Doppler effect results from the thermal motion of target nuclei. This motion causes an effective increase in the width of the resonance as well as a change in the shape.

The cross section for non stationary atoms depends on the relative energy between the neutron and the target nucleus. For a given incident neutron energy this value is obtained by averaging the cross section over the thermal distribution of velocities of the target nuclei. Application of the Maxwellian Distribution for this purpose has been made, but, of course, this is not entirely valid for solids or liquids.

The effects of crystalline binding on the resonance shape was first treated in 1939 by Lamb.⁴ Applying the methods of quantum mechanics, Lamb considered the Doppler distortion for a solid absorber under three types of lattice binding: weak, intermediate

¹D.J. Hughes and R.B. Schwartz, Neutron Cross Sections, BNL Report 325, 2nd Ed. (1958).

²J. Blatt and V. Weisskopf, Theoretical Nuclear Physics, John Wiley and Sons, New York (1952).

³G. Breit and E. Wigner, Phys.Rev. 49, 519 (1936).

⁴W.E. Lamb, Phys. Rev. 55, 190 (1939).

and strong. For the case of strong binding Lamb finds that the level shape has no Doppler broadening. For weak binding the theory shows that the thermal motion of the atoms may be represented as an ideal gas where in place of the real temperature of the sample an effective temperature is substituted. In general the solid absorbers of interest to neutron resonance spectroscopy fall into the category of intermediate binding. For this case the effects of lattice binding distort the resonance shape in a manner that is not simple to express.

Two additional methods for interpreting the effects of the thermal motion of atoms on the shape of neutron absorption lines have been presented recently. One method is by Egelstaff and Holt⁵ and the other by Nelkin and Parks.⁶ Both start their analysis in the same way as Lamb, but their subsequent developments proceed differently. They both show that due to the independence of atomic and nuclear motions, the Doppler broadened cross section can be expressed in terms of the formalism developed for the theory of inelastic scattering of slow neutrons.

The Doppler effect is important in nuclear theory because it distorts the natural resonance shape, and therefore limits the accuracy to which the parameters describing the resonance can be obtained. Greater precision in determining these values can be

⁵P.A. Egelstaff and G. Holt, Inelastic Scattering of Neutrons in Solids and Liquids, I.A.E.A., Chalk River, Vol. II, 183 (1962).

⁶M. Nelkin and D.E. Parks, Phys.Rev. 119, 1060 (1960).

achieved when a better understanding of the thermal motion of atoms is obtained.

The moderation of fission neutrons to thermal energies is one of the fundamental processes that must be considered in reactor design. During this slowing down process it is possible for neutron absorption to take place in capture resonances. Since the Doppler effect causes the resonance absorption cross section to be a function of temperature, the phenomenon of Doppler broadening is of particular importance in reactor technology.

An observed resonance is a superposition of its natural shape with a Doppler shape that depends on the thermal motion of the atoms composing the sample. In view of this dependence it would appear that for a resonance whose nuclear properties were well known, one should be able to extract from the Doppler distortion information about lattice frequency spectra. Since the natural widths of these levels are generally large, it is doubtful, however, that this method for determining lattice spectra would be very sensitive.

B. Scope of This Study:

In selecting a neutron resonance for a Doppler broadening study an important requirement is that the level should have a narrow width. In general a narrow resonance is more severely Doppler distorted than a wide resonance, and therefore presents a more challenging situation for testing the various theories.

The 4.14 ev resonance in tungsten-182 is a narrow resonance and was selected for the Doppler study reported here.

The shape of this resonance was measured to high accuracy at four different temperatures, 4.2°K , 77.8°K , 296°K , and 825°K using a metal sample. The shape of the resonance was also measured using a heavy water solution of sodium tungstate at 296°K .

At a series of intermediate temperatures between 4.2°K and 825°K the peak cross section of the resonance was measured.

To make the measurements above room temperature an oven was used. To make the measurements below room temperature a cryostat was used.

The source of neutrons used in this experiment was the graphite reactor at Brookhaven National Laboratory and the monochromator employed was a crystal spectrometer.⁷

⁷L.B. Borst and V.L. Sailor, Rev. Sci. Instr. 24, 141 (1953); V.L. Sailor, H.L. Foote, H.H. Landon, and R.E. Wood, Rev. Sci. Instr. 27, 26 (1956).

CHAPTER II

THE MATHEMATICAL DESCRIPTION OF NEUTRON RESONANCES

A. The Form of the Cross Section Before Broadening:

1. The Single Level Formula

The two important processes for the interaction of low energy neutrons with intermediate or heavy nuclei are neutron capture and elastic scattering. In the case of neutron capture the compound nucleus decays by the emission of photons, and for elastic scattering the compound nucleus re-emits a neutron with the energy of the incident neutron. For a single isolated resonance the mathematical descriptions for the energy dependence of these processes are given by the Breit-Wigner formulae.

In the laboratory system the equation for the radiative capture of s-wave neutrons close to the resonance energy is given by

$$\sigma_{m,\gamma}(E) = \sqrt{\frac{E_0}{E}} \frac{\sigma_{m,\gamma_0} \Gamma^2}{4(E-E_0)^2 + \Gamma^2} \quad (2.1a)$$

where

$$\sigma_{m,\gamma_0} = \frac{4\pi\lambda_0^2 g \Gamma_m \Gamma_\gamma}{\Gamma^2} \left(\frac{A+1}{A} \right)^2 \quad (2.1b)$$

The symbols used above are now defined:

- E is the kinetic energy of the incident neutron in the laboratory coordinates,
- E₀ is the resonance energy in the laboratory coordinates. All quantities with the subscript '0' are evaluated at E₀.

$\sigma_{m,\gamma}$ is the neutron capture cross section,

σ_{m,γ_0} is the peak value when $E = E_0$,

λ is the neutron's de Broglie wavelength divided by 2π ,

Γ_m is the scattering width of the resonance,

Γ_γ is the capture width of the resonance,

$\Gamma = \Gamma_m + \Gamma_\gamma$ is the total width of the resonance,

g is the statistical weight factor $\frac{2J+1}{2(2I+1)}$, where
 I is the spin of the target nucleus and $J = I \pm \frac{1}{2}$
is that of the compound nucleus,

A is the ratio of the nuclear mass to the neutron mass.

The elastic scattering cross section is composed of three factors: (a) a resonance term, (b) an interference term, and (c) a potential term. The equation for scattering is given by

$$\sigma_{m,m}(E) = \frac{\sigma_{m,m_0} \Gamma^2}{4(E-E_0)^2 + \Gamma^2} + \frac{Q(E-E_0)}{4(E-E_0)^2 + \Gamma^2} + \sigma_p \quad (2.2a)$$

resonance interference potential

where $\sigma_{m,m_0} = \frac{4\pi \lambda_0^2 g \Gamma_m^2}{\Gamma^2} \left(\frac{A+1}{A} \right)^2 \quad (2.2b)$

and $Q = 16\pi \lambda_0 g \Gamma_\gamma \Gamma_m \left(\frac{A+1}{A} \right) \quad (2.2c)$

On the right hand side of Eq. (2.2a) the first term represents the elastic scattering cross section due to the re-emission of a neutron and the last term gives the "hard sphere" contribution. Since these scattering processes are coherent an additional term

arises which is called the interference term. The middle term in Eq. (2.2a) represents the interference between the resonance and potential scattering. R_g is the nuclear radius for the resonant state.

In Eqs. (2.1b), (2.2b) and (2.2c) the $\left(\frac{A+1}{A}\right)$ factors result when the Breit-Wigner formulae are expressed in the laboratory coordinate system. Oftentimes they have been omitted from the analysis of neutron resonances. For nuclei with mass ratios greater than 140, an error of about one percent is introduced, which is usually less than the accuracy quoted for resonance parameters.

The total cross section, σ_{mT} , is given as the sum of the capture and scattering terms, i.e.

$$\sigma_{mT}(E) = \sigma_{m,\gamma}(E) + \sigma_{m,n}(E) \quad (2.3)$$

and it is this quantity which is observed in a transmission measurement.

The determination of resonance parameters that fit the Breit-Wigner single-level formulae has been given a great deal of attention. Its validity has been tested many times and in all cases the experimental data were well matched to within the accuracy of the experiment. From total cross section measurements made in different regions of the resonance curve one is able to determine all the level parameters with two exceptions, namely g and Γ_m . Neither of these two parameters can be obtained individually with any degree of accuracy from total cross section data but are only obtained as the product $g\Gamma_m$. It is to be noted that since W^{182} is an even-even nucleus, its spin

is zero and hence its statistical weight factor, g , is unity. Consequently all the resonance parameters can be determined solely from total cross section data.

B. Derivation of the Usual Form of Doppler Broadening:

1. The Ideal Gas Model

The cross section formulae given in the previous section must now be modified to allow for the thermal motion of the target nuclei. The first to call attention to the Doppler effect in the case of neutron resonances were Bethe and Placzek⁸ in 1937 where the atoms were treated as an ideal gas. They calculated the cross section that would result as a convolution of the natural line-shape with the Maxwellian Distribution of velocities.

Recently the equation which describes the Doppler broadened cross section of target atoms having a Maxwellian Distribution of velocities has been considered in great detail by several authors.^{9,10,11} Their essential modification is to replace the Maxwellian averaged value with an effective cross section which leads to the correct reaction rate. Of course since the cross sections actually measured is a reaction rate per incident neutron and since the resonance parameters reported have their greatest applicability in the calculation

⁸H.A. Bethe and G. Placzek, Phys. Rev, 51, 450 (1937);
H.A. Bethe, Revs. Modern Phys., 9, 69 (1937).

⁹G.I. Bell, Theory of Effective Cross Sections, LA-2322 (Oct., 1959).

¹⁰A.W. Solbrig, Jr., Am. J. Phys., 29, 257 (1961);
A.W. Solbrig, Jr., Nuclear Sci. and Eng., 10, 167 (1961).

¹¹G.W. Hinman and J.D. Sampson, The Rigorous Determination of the Doppler Broadening of Nuclear Resonances for a Maxwellian Gas Absorber, G.A.-3603 (Oct., 1962).

of reaction rates then it follows that this is the more logical way in which to evaluate the Doppler effect.

The effective Doppler broadened value is given by the relation¹²

$$\sigma_{m,x}(\Delta, \bar{v}) = \frac{1}{\bar{v}} \int \bar{v}_n \sigma_{m,x}(\bar{v}_n) w(\bar{v}') d^3 \bar{v}' \quad (2.4a)$$

where $\bar{v}$ is the velocity of the incident neutron,
 $\bar{v}'$ is the velocity of the target nucleus,
 $\bar{v}_n = \bar{v} - \bar{v}'$ is the velocity of the incident neutron relative to the target nucleus, and

$\sigma_{m,x}$ refers to the resonance cross section component with x designating either γ , corresponding to absorption, or m , corresponding to scattering.

w is the Maxwellian velocity distribution,

$$w(v') = \left(\frac{M}{2\pi kT} \right)^{3/2} \exp \left[- \frac{M}{2kT} (v')^2 \right] \quad (2.4b)$$

where M is the mass of the target nucleus,
 k is Boltzmann's constant, and
 T is the temperature.

The inclusion of the relative velocity factor into the integrand and the incident neutron velocity divisor outside the integral in

¹² The Doppler width, Δ , when used as a variable denotes quantities that differ from the theoretical only in that they have been Doppler broadened.

Eq. (2.4a) are essential modifications over the Bethe-Placzek formulation.

With the substitution of Eq. (2.4b) into Eq. (2.4a) and transforming to polar coordinates in the relative velocity space and performing the necessary angular integration, one finds that this equation may be written as

$$\sigma_{m,x}(\Delta, E) = \frac{1}{2} \sqrt{\frac{M}{\pi m k T}} \frac{1}{E} \int_0^{\infty} dE_n \sqrt{E_n} \sigma_{m,x}(E_n) \left\{ \exp\left[-\frac{M}{m k T} (\sqrt{E} - \sqrt{E_n})^2\right] - \exp\left[-\frac{M}{m k T} (\sqrt{E} + \sqrt{E_n})^2\right] \right\} \quad (2.4c)$$

where m is the mass of the neutron,

$$E = \frac{1}{2} m v^2 \quad (2.4d)$$

and

$$E_n = \frac{1}{2} m v_n^2. \quad (2.4e)$$

It is important to note that Eq. (2.4c) is rigorous and does not include the Bethe approximation that the kinetic energy of the neutron is much greater than the kinetic(thermal) energy of the target nucleus.

Further simplification in Eq. (2.4c) is possible. Note that the maximum for the second term in the brace is $\exp\left[-\frac{M}{m} \frac{E}{k T}\right]$. For resonances above $k T$ appearing in nuclei with mass ratios greater than 100, this second term is always less than e^{-100} and may be neglected¹³. Therefore Eq. (2.4c) reduces to

$$\sigma_{m,x}(\Delta, E) = \frac{1}{\sqrt{\pi}} \frac{1}{\Delta} \frac{1}{\sqrt{E}} \int_0^{\infty} dE_n \sqrt{E_n} \sigma_{m,x}(E_n) \exp\left[-\frac{4E(\sqrt{E} - \sqrt{E_n})^2}{\Delta^2}\right] \quad (2.4f)$$

¹³ For resonances below $k T$ this approximation may introduce serious errors. The proper form of the integral as given in Eq. (2.4c) should be used.

where the Doppler width, Δ , has the form

$$\Delta = 2\sqrt{\frac{m}{M} k T E} \quad (2.4g)$$

When a binomial expansion of $\sqrt{E_n}$ is made about $\sqrt{E}$, then

$$\sigma_{m,x}(\Delta, E) = \frac{1}{\sqrt{\pi}} \frac{1}{\Delta} \int_0^{\infty} dE_n \sqrt{E_n} \sigma_{m,x}(E_n) \exp\left[-\frac{(E_n - E)^2}{\Delta^2}\right] \quad (2.4h)$$

which is the form we now wish to use. Substituting Eq. (2.1a) into Eq. (2.4h) where $x = \gamma$; and identifying E and E_0 of Eq. (2.1a) with E_n and E of Eq. (2.4h), respectively, gives the Breit-Wigner Doppler broadened cross section as

$$\sigma_{m,\gamma}(\Delta, E) = \frac{\sigma_{m,\gamma_0}}{\sqrt{\pi}} \frac{1}{\Delta} \sqrt{\frac{E_0}{E}} \int_0^{\infty} dE_n \frac{\exp\left[-\frac{(E_n - E_0)^2}{\Delta^2}\right]}{\left(\frac{E_n - E_0}{\Gamma/2}\right)^2 + 1} \quad (2.4i)$$

Introducing certain non-dimensional variables as

$$x = \frac{2(E - E_0)}{\Gamma} \quad (2.4j)$$

$$y = \frac{2(E_n - E_0)}{\Gamma} \quad (2.4k)$$

and
$$t = \left(\frac{\Delta}{\Gamma}\right)^2 \quad (2.4l)$$

Eq. (2.4i) becomes

$$\sigma_{m,\gamma}(\Delta, E) = \sigma_{m,\gamma_0} \sqrt{\frac{E_0}{E}} \psi(x, t) \quad (2.4m)$$

where $\psi(x, t)$ is what we shall call the Doppler integral¹⁴

¹⁴

Westinghouse Atomic Power Division Report SR-506 (1954).

$$\Psi(x,t) = \frac{1}{2\sqrt{\pi t}} \int_{-\infty}^{+\infty} \frac{\exp\left[-\frac{(x-y)^2}{4t}\right]}{1+y^2} dy \quad (2.4n)$$

An analogous treatment shows that when the Breit-Wigner scattering cross section, where $\chi = m$, Eq. (2.2b), is substituted into Eq. (2.4h) the resultant expression is given by

$$\sigma_{m,m}(\Delta, E) = \sigma_{m,m_0} \Psi(x,t) + \frac{2}{2\Gamma} \phi(x,t) + \sigma_p \quad (2.5a)$$

resonance

interference

potential

$\phi(x,t)$ is another integral¹⁵ given by

$$\phi(x,t) = \frac{1}{2\sqrt{\pi t}} \int_{-\infty}^{+\infty} y \frac{\exp\left[-\frac{(x-y)^2}{4t}\right]}{1+y^2} dy \quad (2.5b)$$

In the evaluation of the scattering terms a factor under the integral of $(E/E_0)^{1/2}$ has been set equal to unity. For many resonances the scattering contribution to the total cross section is small and this approximation can be assumed to apply in these cases to high accuracy.

According to this development the total broadened resonance cross section for an ideal gas target is the sum of Eq. (2.4m) and Eq. (2.5a), i.e.

$$\sigma_{mT}(\Delta, E) = \left(\sqrt{\frac{E_0}{E}}\right) \sigma_{m,\chi_0} + \sigma_{m,m_0} \Psi(x,t) + \frac{2}{2\Gamma} \phi(x,t) + \sigma_p \quad (2.6)$$

¹⁵

M. Born, Optik, p. 486, Springer, Berlin (1933).

C. Derivation of Doppler Broadening for a Solid Absorber :

1. The Theory of Lamb⁴

Target materials used in neutron resonance spectroscopy cannot always be obtained in a gaseous form, e.g. pure tungsten. Therefore measurements of total cross sections must generally rely on samples where the target atoms are in a solid or in a liquid solution.

The theory of neutron capture by atoms bound in a crystal was first considered by Lamb. In his theory Lamb considered the effects of lattice binding on the shape of a pure capture resonance. A great deal of interest in Lamb's work has been stimulated in recent years due to the discovery of the Mössbauer Effect.¹⁶ In Mössbauer's theory of recoilless emission and absorption of gamma rays, the starting point that he used for interpreting the recoilless effect was the theory originally presented by Lamb many years ago.

Lamb treats the solid absorber as a quantum mechanical system and develops his theory in terms of the formalism of quantum mechanics. Since lattice forces are known to be weak they are not able to noticeably affect the motions of the nucleons inside the nucleus. In view of this Lamb was able to separate the process of compound nucleus formation from the lattice effects. These separated processes, however, are related to each other from a consideration of the applicability of the conservation laws. The energy and momentum associated with the recoiling of the compound nucleus in formation as well as in its decay must appear in the changes in the states of the lattice vibrations.

⁴Lamb, op.cit.

¹⁶R.L. Mössbauer, Z. Physik, 151, 124 (1958); Naturwiss., 45, 538 (1958).

One of the simplifying assumptions made by Lamb is that the theory deals with simple and isotropic crystals. In normal coordinates such a crystal can be treated as a system of harmonic oscillators. According to this theory the line shape that results for a pure capture resonance is given by

$$\sigma_{m,r}(\Delta, E) = \frac{\sigma_{m,r_0} \Gamma^2}{4} \operatorname{Re} \int_0^\infty du e^{i u (E - E_0 + \frac{i\Gamma}{2})} g(u) \quad (2.7a)$$

where

$$g(u) = \sum_s g_s^2 \left[(\bar{\alpha}_s + 1) e^{-i u \hbar \omega_s} + \bar{\alpha}_s e^{i u \hbar \omega_s} - 1 - 2 \bar{\alpha}_s \right]. \quad (2.7b)$$

Here ω_s is the frequency of the s^{th} normal mode, g_s is the wave propagation vector, and $\bar{\alpha}_s$ is the number of phonons in the s^{th} mode of oscillation. An explanation of Lamb's notation and an outline of his theory are given in Appendix A.

It is not possible to reduce $\sigma_{m,r}(\Delta, E)$ further because of the complexity of $g(u)$ in the exponential. Lamb, however, does consider two limiting cases: (a) the weak binding approximation where $u \hbar \omega_s \ll 1$, and (b) the strong binding approximation where $u \hbar \omega_s \gg 1$.

In the weak binding limit E. (2.7a) reduces to

$$\sigma_{m,r}(\Delta, E) = \sigma_{m,r_0} \Psi(x, t) \quad (2.7c)$$

where $\Psi(x, t)$ is the same integral given in Eq. (2.4n). Therefore the shape of the broadened line for a solid absorber is the same as for the ideal gas absorber. There is a difference, however, which

will now be explained. The difference is in a new definition of the Doppler width, Δ . For the weak binding crystal Δ depends on the average energy per degree of vibration in place of the sample's temperature as given in Eq. (2.4g).

A familiar way to describe the properties of a crystal is in terms of the Debye model. According to this model the velocity of sound waves (phonons) is assumed independent of wavelength, direction and polarization. From its success in describing specific heats, the Debye model is known to be a fair approximation for effects which involve the average of a smooth function over all crystal vibrations. Applying this model, Lamb was able to relate the average energy per degree of vibration of the crystal to an effective temperature, T' , which is to be used in place of the sample's real temperature, T . Therefore knowing the Debye temperature of a material one is able to determine the sample's effective temperature by the following relation¹⁷

$$\frac{T'}{T} = \frac{C_v(\theta/T)}{24} + \frac{3(\theta/T)}{4[\exp(\theta/T) - 1]} + \frac{3(\theta/T)}{8} \quad (2.7d)$$

where C_v is the atomic heat function.¹⁸ Many known resonances

¹⁷

J.E. Lynn and E.R. Rae, J. Nucl. Energy, 4, 418 (1958);

In the report LA-2763, Hafnium Cross Sections and Their Temperature Dependence, authors J. Devaney et al. indicate that the divisor, 24, in Eq. (2.7d) should be replaced by 12. Our results indicate that Eq. (2.7d) is correct. If this change were made we observe that this form would not then agree with the alternate expansion given in LA-2763 in footnote 4. Eq. (2.7d) would agree, however.

¹⁸

For tables of C_v see Roberts and Miller, Heat and Thermodynamics, 5th ed., p537, Interscience, New York (1951).

have been analyzed for their Breit-Wigner parameters using this weak binding limit and it has become popularly known as the effective temperature model.

In the strong binding limit Lamb finds that the resonance shape has the pure Breit-Wigner dependency indicating that there is no Doppler broadening. In this case the resonance energy, E_0 , is unshifted. In the weak binding limit the line form is not centered about the resonance energy but is shifted to a higher energy given by

$$E = E_0 + R \quad (2.7e)$$

where R is the free atom recoil energy.

When the solid absorber is treated as a Debye crystal in the weak binding limit, Lamb has shown that the requirement $u \hbar \omega_s \ll 1$ is equivalent to the condition that

$$\Gamma + \Delta \gg 2\Theta \quad (2.7f)$$

Here Γ is the resonance's natural width as previously defined,

Δ is the effective Doppler width for the crystal, and Θ is the sample's Debye temperature expressed in energy units. For most resonance cases the weak binding condition given in Eq. (2.7f) is too vague and a practical limit to the inequality is preferred. Egelstaff¹⁹

¹⁹P.A. Egelstaff, A.E.R.E. Report M-1035(1962).

has investigated this requirement by performing shape calculations on the 6.65 ev resonance in U-238. Here Θ was permitted to vary but the temperature of the sample as well as the values for Δ and Γ were kept constant. Thus the effect studied was the change in the shape of the resonance with a change of Debye temperature, i.e. the strength of the chemical binding. These results indicate that the ratio $\frac{\Gamma + \Delta}{2\Theta}$ need only be greater than approximately 2 before the weak binding model can be applied confidently.

In the use of Lamb's results it is important to remember that this theory applies only to resonances that are predominantly capture and is not applicable to scattering resonances.

D. Application of Inelastic Neutron Scattering Theory to Resonance Analysis:

1. The Theory of Egelstaff and Holt⁵

A modern formula that expresses the Doppler broadened cross section for a capture resonance has been given by Egelstaff and Holt. Beginning with formula (26) of Lamb⁴ (see also Appendix A, Eq. (A.21a)), Egelstaff has shown²⁰ that the resonance formula takes the form

$$\sigma_{n,\gamma}(\Delta, E) = \frac{\sigma_{n,\gamma_0} \Gamma^2}{4} \int_{-\infty}^{+\infty} \frac{\exp\left[-\frac{\beta}{2}\right] S_s(\alpha, \beta)}{(E - E_0 - \beta kT)^2 + \left(\frac{\Gamma}{2}\right)^2} d\beta. \quad (2.8)$$

Thus the observed shape reduces to a convolution of the Breit-Wigner capture part with the "self" part of a Scattering Law, $S_s(\alpha, \beta)$, which can be obtained from inelastic neutron scattering measurements. Physically one understands this as a result of the independence of nuclear and atomic motions.

⁴Lamb, op. cit.

⁵Egelstaff and Holt, op. cit.

²⁰P.A. Egelstaff, private communication.

In recent years a great deal of research has been done in the field of inelastic scattering of slow neutrons in solids and liquids.²¹ The reason for this activity is that these measurements are capable of yielding information on the nature of interatomic forces not attainable previously.²²

In these experiments a beam of monoenergetic neutrons is scattered by a fixed target and those neutrons that are inelastically scattered are then measured to determine their energy distribution. When the scattering is inelastic both energy and momentum are exchanged between the neutron and the scatterer satisfying conservation requirements. The dispersion curves obtained may then be analyzed to determine the nature of the interatomic forces.

The intensity distribution for the scattered neutrons is given in terms of the differential cross section, $\frac{d^2\sigma}{d\Omega dE}$, which expresses the probability that a neutron with an initial energy E scatters into an element of solid angle $d\Omega$ having a final energy E' . In this process the energy gained by the neutron is $\epsilon = E' - E$. For an isotropic scatterer Egelstaff and Schofield²³ find it convenient to write the cross section obtained by Van Hove²⁴ as

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b}{4\pi kT} \left(\frac{E'}{E} \right)^{\frac{1}{2}} \exp\left(-\frac{\beta}{2}\right) S(\kappa, \beta) \quad (2.9a)$$

²¹Proceedings on Inelastic Scattering of Neutrons in Solids and Liquids, I.A.E.A., Vienna (1961) and Chalk River (1963); Proceedings of the Brookhaven Conference on Neutron Thermalization, BNL Report 719, (1962).

²²With X-rays one cannot get high enough resolution to measure energy changes in the scattered beam, while with infrared radiation one can see only atomic motions in which very large numbers of atoms move together as a group. With slow neutrons, however, one is able to see almost any energy change that is likely to occur.

²³P.A. Egelstaff, Nuclear Sci. and Eng. 12, 250(1962); P.A. Egelstaff and P. Schofield, Nuclear Sci. and Eng. 12, 260 (1962).

²⁴L. Van Hove, Phys. Rev. 95, 249 (1954).

where σ_b is the bound atom cross section
 k is the Boltzmann constant, and
 T is the absolute temperature of the scatterer.

$S(\alpha, \beta)$ is called the Scattering Law and it is a property of the scatterer. By application of Eq. (2.9a) all measurements yielding the differential scattering cross section may be reduced to this Scattering Law. S is a function of two variables only, α and β . α is the momentum transferred in the scattering and it is expressed in dimensionless units as

$$\alpha = \frac{1}{A k T} \left[E' + E - 2 (E E')^{1/2} \cos \theta \right] \quad (2.9b)$$

where θ is the scattering angle. β is the energy transferred also in dimensionless units,

$$\beta = \frac{1}{k T} (E' - E) \quad (2.9c)$$

For the case of a perfectly inelastic collision where the incident neutron disappears, e.g. compound nucleus formation, then $E' = 0$. A property of $S(\alpha, \beta)$ is that it is an even function of β . This is because it is equally probable that the neutron will either gain or lose energy upon being scattered.

In order to apply the results of inelastic scattering measurements to resonance analysis, the Scattering Law must be corrected first for interference effects. The function S may be divided into two parts: a self term, S_s , which gives the scattering by a single nucleus, and an interference term which results from waves scattering from several

atoms. The procedure for obtaining S_s from experimental measurements has been considered by Egelstaff.²³

To assist in the evaluation of the Doppler broadened shape by this procedure, R.C.F. McLatchie of Harwell has written two computer programs.²⁵ For any assumed phonon frequency spectrum, the function

S_s is first calculated by program LEAP. The second program, called LINESHAPE, evaluates numerically the integral of Eq. (2.8). Of course for a particular resonance values for $\sigma_{m,0}$ and Γ must be supplied.

Egelstaff and Holt⁵ have used this method to calculate transmission curves for several different frequency distributions. The parameters for the 4.26 ev resonance in tantalum were used in this study. It is interesting to observe that for the distributions assumed small differences in the transmission can be seen. However, it would appear that even in the case of an accurate experiment to measure the shape of a resonance the determination of a unique frequency spectrum by this method is doubtful.

2. The Theory of Nelkin and Parks.⁶

Another formula expressing the Doppler broadened cross section for a capture resonance has been given by Nelkin and Parks. In this

⁵ Egelstaff and Holt, op. cit.

⁶ Nelkin and Parks, op. cit.

²³ Egelstaff, op. cit.

²⁵ These elaborate programs have been made available to many research groups.

development the formalism used in slow neutron scattering is again applied. Starting with formula (11) of Lamb⁴ (see also Appendix A, Eq. (A.10)) and making asymptotic expansion for large nuclear recoil it is found that the Doppler broadened line shape is given by the formula

$$\sigma_{m,\gamma}(\Delta, E) = \sigma_{m,\gamma_0} \Psi'(x, t) \quad (2.10a)$$

where

$$\Psi'(x, t) = \Psi_0(x, t) + \sum_{m=3}^{\infty} A_m \Psi_m(x, t) \quad (2.10b)$$

and

$$\Psi_m(x, t) = \frac{1}{2\sqrt{\pi t}} \int_{-\infty}^{+\infty} \frac{H_m\left(\frac{x-y}{\sqrt{2t}}\right) \exp\left[-\frac{(x-y)^2}{4t}\right]}{1+y^2} dy. \quad (2.10c)$$

The leading term of the expansion, Ψ_0 , in Eq. (2.10b) is the same as the weak binding limit obtained by Lamb (see Appendix A, Eq. (A.28)).

H_m is called the m^{th} order Hermite polynomial. These polynomials however, must be obtained by use of the generating function given by the authors. Their generating function differs from the one usually employed to give these polynomials.

The coefficients, A_n , have been evaluated for a Debye crystal model and formulas for them are given in the report.⁶ An inspection of them reveals that the higher terms of the expansion become increasingly important as the Debye temperature of the absorber increases or as the free atom recoil energy or the physical temperature of the sample decreases.

A mathematical condition stating when this expansion approximation may be applied is not given. The "large recoil" limit is too

⁴Lamb, op. cit.

⁶Nelkin and Parks, op. cit.

vague to be useful. It would appear that this method should be applied subject to the same requirement as Lamb's weak binding limit.

E. Doppler Broadening for an Absorber in a Liquid:

In contrast to the gaseous and the solid states of matter, the dynamics of the liquid state are not as well understood. The atomic and molecular motions in liquids are very complex and there does not exist any satisfactory theory for this state.²⁶

Resonance studies performed where the target was a liquid specimen, e.g. some chemical compound including the target atom dissolved in heavy water, have been analyzed for the Doppler effect in terms of the ideal gas model. This procedure has proven to be adequate. Adequate here is interpreted to mean that the resonance curve can be fitted to within the errors of observation by a unique choice of Breit-Wigner parameters. Of course realizing that there are a relatively large number of parameters involved in the fitting does not make this result surprising. If, however, the shape of a level were determined to very high accuracy, it is not certain that this same conclusion would be observed.

F. Additional Broadening Caused by Instrumental Resolution:

Implicit in the treatment of the Doppler effect as it has been considered thus far is the assumption that the neutron beam incident on the assembly of thermally moving atoms is purely monoenergetic with energy E . For this ideal situation the transmission, T , through

²⁶ Experiments on liquids using the inelastic scattering of neutrons technique indicate that this method is a powerful tool for determining some of the fundamental properties of liquids. Work on this problem is in progress at many of the research centers throughout the world. Conclusive interpretation of these results, however, will require a great deal of effort. The theoretical formalism used in the analysis of the data is usually that of Van Hove.

the sample is given by

$$T(\Delta, E) = \exp \left[-N \sigma_{\text{MT}}(\Delta, E) \right] \quad (2.11)$$

where N is the number of target atoms per cm^2 , and σ_{MT} is the total Doppler broadened cross section.

In practice, however, it is not possible to achieve this condition of a monoenergetic beam. For a Bragg diffracted beam obtained with a crystal spectrometer, the neutrons are distributed over a small range of energies due to the resolution of the system. The general form of the distribution for a crystal spectrometer⁷ is given by the function

$$Q(E'-E) = A E'^{-\frac{5}{2}} \exp \left[-\frac{4 \ln 2}{(\Delta E)^2} (E'-E)^2 \right] \quad (2.12)$$

Q is called the resolution function when the energy setting of the spectrometer is E . E' is the variable energy at which Q is evaluated and A is a numerical constant. The quantity ΔE is the width of the resolution function at half maximum and its value is characteristic for a given monochromator.

An observed transmission measurement made at the nominal energy E , is expressed by the relation

$$T_{\text{obs}}(E) = \int_{-\infty}^{\infty} T(\Delta, E') Q(E'-E) dE' \quad (2.13)$$

where Q is assumed to be normalized. Thus all observed transmission are convolutions of Doppler broadened transmissions with the resolution function of the instrument used in the measurement. Interpretation of this in terms of the resonance shape means that the level is further broadened and that Doppler effects cannot be observed directly.

⁷Sailor et al., op. cit.

CHAPTER III

EXPERIMENTAL EQUIPMENT

A. The Neutron Spectrometer:

1. General Information

The source of neutrons used in this research was the graphite reactor located at Brookhaven National Laboratory, Upton, N.Y. Neutron resonance studies designed to obtain level parameters require, in general, the use of neutrons with well defined kinetic energies. A neutron beam emerging from a reactor, however, possesses energies ranging from several Mev down to the thermal region. The monochromator used to obtain a monoenergetic beam from the "raw" reactor spectrum was a crystal spectrometer presently in operation at Brookhaven. A detailed description of this spectrometer has been given before.⁷ In the following sections only the general features of the equipment are reviewed.

A neutron spectrometer is an apparatus which uses the coherent scattering of neutrons from the nuclei of atoms in a single crystal to produce a monoenergetic neutron beam. In use, the crystal is placed so that a reflection is obtained in a certain direction from a particular plane. Its principle of operation is similar to a Bragg X-ray spectrometer. The scattering of neutrons into a reflected beam occurs only for those neutrons whose de Broglie wavelength, λ ,

⁷ Borst and Sailor, op. cit.

satisfies the Bragg relation

$$n \lambda = 2 d \sin \theta \quad (3.1)$$

where $n = 1, 2, 3, \dots$ is the order of the reflection, d is the spacing of the appropriate crystal planes, and θ is the angle between the incident beam and the reflecting planes. Since all order of neutrons above the primary energy of the reflected beam also satisfy the Bragg condition, a correction is introduced in the processing of transmission data to account for these higher energy components.

Different energies of the Bragg reflected monoenergetic beam are obtained by variations in the angle, θ . The angular movements of arm and crystal table is fixed at two in order that the reflected beam should always enter the BF_3 detector located at the end of the spectrometer arm. In order to insure that the incident beam intensity remains constant during a counting run, a small BF_3 monitor records neutrons scattered off of the first Soller collimator in such a way as not to interfere with the beam incident upon the crystal. All data recorded by the main counter are normalized to the monitor counter for the same time intervals.

A diagram of the BNL reactor-spectrometer complex is shown in Figure 1. The "neutron source" indicated in this diagram is a block of graphite placed in the center of the reactor which scatters part of the neutron flux in the direction of the port where the spectrometer is located. At the center of the reactor the neutron flux is estimated to be approximately 2×10^{13} thermal neutrons/cm²/sec when the reactor is running at its full normal power of 20 MW.

**CRYSTAL SPECTROMETER
FOR NEUTRON CROSS SECTION MEASUREMENTS
(SCHEMATIC)**

Fig. 1 Schematic diagram of the reactor-spectrometer complex.

2. Beryllium Monochromator

The effectiveness of a neutron spectrometer as a grating for producing a monochromatic neutron beam from the pile spectrum depends strongly on the crystal used. One of the preferred choices of crystals for this application is beryllium. Its large coherent scattering cross section of 7.54 barns as well as its small thermal absorption cross section, 10 millibarns, make it suitable for this purpose. Beryllium has a hexagonal, close-packed structure so that its interatomic spacings are small. Small grating space and high coherent scattering cross section indicate that (a) a considerable number of suitable planes are available, (b) the useful energy range is large, (c) the energy resolution is high, (d) the reflectivity from a set of planes is strong, and (e) the order discrimination can be enhanced.

In this experiment the $\text{Be}(12\bar{3}1)$ planes were used as monochromator because experience has shown them to be a good compromise between intensity and resolution. The size of the single beryllium crystal used is exceptionally large, 6 in. x $2\frac{1}{2}$ x $\frac{1}{4}$ in., and it has a rocking curve width of 3.7 minutes. Additional advantages to using these crystal planes are: (a) the second order reflection is suppressed 3:1 because of the crystal structure factor and 4:1 because of the Debye-Waller factor, and (b) because of the crystal structure factor for third order reflection is zero, i.e. forbidden.

3. Collimation System

In addition to the crystal limitations, a neutron spectrometer does not have perfect resolution because of the angular divergence of the collimating system. To minimize this condition the beam is

required to pass through two identical Soller collimators, one is located in the reactor shield and the other on the spectrometer arm. These collimators are each 48 inches long and they are placed symmetrically about the crystal. Each collimator has a divergence of only 2 minutes of arc.

For the $\text{Be}(12\bar{3}1)$ planes and for the collimation system installed, the resolution of the spectrometer is given by $\Delta E = 0.00474 E^{3/2}$. For the energy of the 4.14 ev tungsten resonance ΔE is equal to 0.040 ev and this is the value used in Eq. (2.12).

4. Energy Calibration

Energy measurements made with a crystal spectrometer are capable of very high accuracy since its construction and adjustment can be accomplished to high precision. In order to know the absolute energy of the reflected beam, however, the absolute value of the Bragg angle, θ , as read from the crystal angular circle must be properly determined. This scale is properly calibrated when its zero reading corresponds to the "head-on" direction of the reactor beam. This direction can be observed easily by moving the counter through the direction of the unreflected beam and observing a maximum in the counting rate.

During the 18 months of data recording required for this experiment the zero angle of the spectrometer was measured several times. Each time the constancy of the beam zero with the zero scale reading was indistinguishable.

B. Target Samples:

1. Metallic Foils

The Doppler effect on the shape of the 4.14 ev resonance was studied in metallic samples of natural tungsten.²⁷ The purity of the samples were 99.9 percent or better and were uniform in thickness to ± 1 percent. Spectrographic analysis made at Brookhaven confirmed their chemical purity. X-ray pictures taken showed uniform exposure indicating a homogeneous distribution and the absence of voids.

Two metal samples of tungsten were used. The sample used to measure the resonance shape at all temperatures had a density of 0.1399 g/cm^2 . In the wings of the resonance where the potential cross section was evaluated the sample had a density of 14.19 g/cm^2 . In units of $1/N$ the thin sample's thickness was 2182 barns/atom and the thick sample was 21.51 barns/atom. These values were determined at 296°K . At each temperature in the range from 4.2°K to 825°K at which cross section measurements were performed on the thin sample, the $1/N$ value for this target was corrected for the effects of thermal expansion. The over all change in this value was 0.7 percent.

2. Liquid Solution of Na_2WO_4

The shape of the resonance was also measured at 296°K where the target was a heavy water solution of sodium tungstate. The material used to prepare the sample was obtained from Baker and Adams and spectrographic analysis indicated no significant impurities.

²⁷ These targets were supplied as a courtesy of the Wah Chang Corporation, Glen Cove, New York, through the kind assistance of Mr. Ned Chou, Chief Metallurgist.

The preparation of the sample was carried out according to standard procedures. The thickness value was 2249 barns/atom. It was early recognized that a shape comparison between a solid absorber and a liquid solution at the same temperature would be simplified if the two samples had approximately the same thickness value. Fortunately this was possible to achieve since sodium tungstate is highly soluble in heavy water. D_2O is preferred to H_2O in order to minimize the scattering of the neutron beam by the solvent. Great care was exercised in the sample preparation so that any error is believed negligible compared to the 0.5 percent uncertainty in the chemical analysis of the Na_2WO_4 . The sample was sealed in a precision quartz cell. Transmission measurements were made against an identical cell filled with heavy water only. To account for the absence of the sodium and oxygen in the D_2O "dummy" cell, a small correction to the observed transmission was required. Measurements made on the D_2O cell revealed that only 64 percent of the incident neutron beam intensity was transmitted.

C. Assemblies for Keeping the Sample Isothermal at Different Temperatures.

Previously it was stated that the shape of the 4.14 eV tungsten resonance has been measured at four different temperatures, namely, 4.2°K, 77.8°K, 296°K and 825°K. The room temperature measurements were made with the sample changer device that is part of the standard equipment of the spectrometer. It is fully automatic and provides for the changing of samples. The measurements made above room temperature were obtained with the use of an oven. The measurements made below room temperature were made with a cryostat. The

Fig. 2 Photograph of spectrometer. The neutron beam emerges from the reactor wall (1). The beryllium crystal is underneath the lucite cover (2). The support column for the oven and cryostat assemblies is shown at (3). The cryostat is visible inside this column. The vacuum pumping station is at (4). The spectrometer arm (5) supports a Soller collimator. It is hidden from view under masonite shielding. The main BF_3 counter is located inside the shield at (6).

photograph (Figure 2) shows the crystal spectrometer used in this experiment. The column shown in this figure as item (3) is mounted to the spectrometer arm. This device can be used to support interchangeably the oven and the cryostat. A drive unit is located on the top of this column which is controlled by the master program panel. By means of this unit the sample can be rotated through an angle of 90 degrees at the proper time in the program. This rotation removes the sample from the incident monoenergetic beam and permits the open beam intensity to be measured. This open beam intensity is necessary in order to calculate the transmission accurately. A microswitch arrangement assures the precision with which this rotation is executed.

1. Oven Assembly

The resonance shape at 825°K was obtained with the apparatus shown schematically in Figure 3. The sample was heated by means of two Vulcan pancake heaters placed on top and bottom of a massive copper sample holder. In this arrangement the sample was sandwiched between the body of the holder and a backing frame that kept the sample firmly against the holder. To minimize heat losses from the oven, the chamber was evacuated to a pressure of 50×10^{-6} mm of Hg. and a series of floating, multiple reflecting radiating shields made of aluminum were inserted. To maintain the sample in thermal equilibrium approximately 70 watts of electrical power were required. With this design the outside wall never exceeded a temperature of 400°C. To protect the spectrometer arm against unnecessary heating, a fan was directed against the oven during the measurements.

OVEN SAMPLE ASSEMBLY

Fig. 3 Schematic diagram of the assembly used for the high temperature measurements. The sample is displaced from the center line of the unit and a 90 degree rotation takes it completely out of the neutron beam. The chamber was evacuated to a pressure of 50×10^{-6} mm Hg. Thermocouple readings were continuously recorded at both the top and bottom of the sample.

Two chrome vs. alumel thermocouples were placed at the top and bottom of the sample. These wires were electrically connected to the terminals of a two-point Leeds & Northrup Speedomax so that a continuous monitoring of the sample's temperature, top and bottom, was obtained as a permanent record. This chart showed that over long periods of time the temperature across the sample never varied more than 10 degrees and often it was much less.

The outside container was also made of aluminum in order to minimize the attenuation of the neutron beam. It was experimentally found that 83 percent of the incident intensity was transmitted through all the Al walls of the oven.

In addition to the resonance shape measurement at 825°K , peak cross section measurements were made also at temperatures at 423°K , 568°K and 709°K by varying the input electrical power to the oven.

2. Cryostat Assembly²⁸

The tungsten foil was maintained at liquid helium temperature, 4.2°K , or liquid nitrogen temperature, 77.8°K , by placing the sample in thermal contact with reservoirs of liquid helium or liquid nitrogen. A schematic diagram for this arrangement is shown in Figure 4. The cryostat's capacity for liquid helium was $2\frac{1}{2}$ liters and it had a staying time of two days without refilling.

The sample holder was held tightly against the plate terminating the tail section of the helium region by 8 screws. Between the sample holder and the plate a layer of Apiezon P grease was placed to enhance the thermal bonding of these two pieces.

²⁸The cryostat was purchased from Superior Air Products, Newark, N.J.

CRYOSTAT SAMPLE ASSEMBLY

Fig. 4 Schematic diagram of the assembly used for the low temperature measurements. The sample is displaced from the center line of the unit and a 90 degree rotation takes it completely out of the beam. The pressure in the vacuum section was approximately 1×10^{-6} mm Hg. Thermocouple readings at both top and bottom of the samples were recorded.

The sample itself was sandwiched firmly between the main section of the sample holder and a backing frame by additional screws.

All materials used in the sample support and in the thermal conduction path were fabricated from oxygen free copper because its coefficient of heat transfer at low temperatures is superior to normal copper.

Two shields were included in the design of the cryostat's sample section to minimize the heat input to the sample from the outside by radiation. The innermost shield was mechanically attached to the helium supply and as such approximates an isothermal enclosure for the sample. The middle shield was attached by screws to the liquid nitrogen jacket which surrounds the helium region. These two radiation shields and the outside container were all made from thin wall aluminum tubing. The neutron transmission through these aluminum cups was measured to be 74 percent of the incident intensity.

The temperature of the tungsten sample was measured at top and bottom with thermocouples. The thermoelectric method for determining temperature was made by the standard procedure using a type K potentiometer. The reference junction was immersed in a liquid nitrogen bath when the sample was at helium temperature, and in an ice-water bath when the sample was at nitrogen temperature.

In order to obtain a large thermoelectric effect at low temperatures a special thermocouple wire was used consisting of gold-2.1 atomic percent cobalt.²⁹ In conjunction with this gold wire, the

²⁹ This precious metal wire was purchased from the Sigmund Cohn Corporation of Mount Vernon, N.Y.

other thermocouple wire used was copper. Calibration tables for the thermoelectric potentials of these wires is given in a report by Powell, Bunch and Corruccini³⁰ of the National Bureau of Standards.

Thermoelectric voltage readings were frequently recorded during the time of transmission measurements by balancing the potentiometer circuit. Over long periods of time the results indicated that the temperature gradient across the sample was less than a degree when the reservoir was filled with liquid helium or liquid nitrogen.

In addition to using the cryostat assembly to measure the resonance shape at 4.2°K and 77.8°K, the peak cross section was also measured at 185°K. The temperature of the sample was reduced to this value by placing a slurry of dry ice and alcohol in the helium section of the cryostat. The temperature reported here is slightly below that listed in the handbook because of the additional cooling from the liquid nitrogen surrounding it.

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R.L. Powell, M.D. Bunch, and R.J. Corruccini, Cryogenics 3, 1 (1961).

CHAPTER IV

THE PROCESSING OF CRYSTAL SPECTROMETER DATA

In this chapter the procedures for obtaining the resonance cross section data from counting rates, the corrections to be applied to the observed cross sections, and the qualitative features of the data are discussed.

A. The Measurement of Total Cross Section:

1. Relation Between Transmission and Cross Section

At any given energy, E , the total cross section can be determined by observing the attenuation of a neutron beam by a sample of known thickness. The observed cross section, σ_{obs} , is given by the relation

$$\sigma_{obs}(E) = \frac{1}{N} \ln \frac{1}{T_{obs}(E)} \quad (4.1a)$$

where N is the areal sample density expressed in atoms/cm², i.e. $N = nt$ where n is the number of target nuclei per cm³ and t is the sample thickness in cm, and

T_{obs} is the transmission ratio of the neutrons which pass through the sample to the incident intensity.

In a resonance study the σ_{obs} value as obtained by Eq. (4.1a) includes effects not directly related to the resonance structure, e.g. environmental background effects, reflected beam contamination with higher order neutrons, sample contaminant with nuclei not under investigation, and cross section contributions from other resonances. Before the resonance shape can be analyzed for level parameters σ_{obs} must first be corrected for these non-resonant effects.

2. Relation Between Counting Rates and Transmission

In addition to the detection of Bragg-reflected neutrons which pass through the sample, non-resonant neutrons also will be counted due to background. Denoting the counting rate with the sample in the beam by C_s , and with the sample out of the beam by C_o , i.e. the open beam count rate, then T_{obs} is given by the relation

$$T_{obs} = \frac{C_s - B_s}{C_o - B_o} \quad (4.1b)$$

where B_s and B_o are the appropriate background counts with the sample in and out of the beam, respectively. Using Eq. (4.1b) to calculate T_{obs} takes due care of the background effects.

3. Statistical Uncertainty in Cross Section

One of the major uncertainties in cross section measurements arises from counting statistics. Assuming that the error in the sample thickness is negligible, i.e. $\Delta N \approx 0$, it can be proved directly by use of Eqs. (4.1a) and (4.1b) that the standard deviations in the transmission and cross section due to statistical uncertainty are given by the relations

$$\Delta T = \frac{1}{C_o - B_o} \sqrt{(C_s + B_s) + T_{obs}^2 (C_o + B_o)} \quad (4.1c)$$

and
$$\Delta \sigma = \frac{1}{N} \frac{\Delta T}{T} \quad (4.1d)$$

In the results reported in this dissertation sufficient counts were taken at each energy that the relative error in the cross section due to counting statistics, i.e. $\frac{\Delta \sigma}{\sigma}$, was one percent or less.

One of the common maladies of experimentation of this sort is the misbehavior of the electronics. Unless particular attention is

paid regularly to the stability of the electronics it is possible for "bad" data to be recorded over an appreciable time interval before the malfunctioning is detected. To prevent such a possible occurrence during this experiment, a surveillance over the electronics was kept by means of Pearson's Chi-square test³¹. This test was performed daily on the open beam measurements of the previous day. In this regard a rule-of-thumb observation was that when the P-value obtained in this test remained low over several consecutive days, it usually fore-warned a subsequent breakdown of one or more major electronic components.

4. Spectrometer Automation

The spectrometer is designed to be fully automatic. In the automation there is a device for putting the sample into the beam as well as taking it out. A crystal shifter device permits counts to be taken with the crystal adjusted for the Bragg condition (termed "on-Bragg") and with the crystal rotated from the Bragg condition by about one degree (termed "off-Bragg") to allow the background counting rates to be measured. With the crystal in the off-Bragg position no Bragg scattered neutrons enter the detector. The master program of the spectrometer permits the "sample-in", open beam and background runs to be taken according to a predetermined plan so that these measurements are interleaved. When the automatic control is switched on it starts a count for a pre-set time interval (usually 20 minutes)

³¹R.D. Evans, The Atomic Nucleus, Chapter 27, McGraw-Hill Book Company, New York (1955).

and this same time duration is used for all measurements.

The spectrometer can be set to record transmission data at different energy settings automatically. However, due to the narrowness of the tungsten resonance and the high accuracy desired in the angular (energy) setting of the spectrometer, all positioning of the spectrometer arm were performed manually.

B. Corrections to the Data:

Before starting the resonance analysis, Chapter V, it is necessary to remove all non-resonant parts of the cross section. These corrections are considered in the following sub-sections.

1. Higher Order Neutrons

In evaluating crystal spectrometer data a problem arises because the reflected beam is not monoenergetic. The spectrometer beam contains neutrons of energy E , $4E$, $9E$, etc., and in accurate work corrections must be made for contamination of the neutron beam by higher order reflections. For studies where the primary energy of the reflected beam is greater than 1 ev, all higher orders amount to only a few percent so that it is found that in most cases all orders except the second (at $4E$) can be ignored. When the $\text{Be}(12\bar{3}1)$ crystal planes are used as monochromator, this approximation is especially good since third order reflections is forbidden by the crystal structure factor.

The observed transmission of a sample placed in the reflected beam is given by

$$T_{\text{obs}} = f_1 T_1(E) + f_2 T_2(4E) \quad (4.2a)$$

where f_1 is the fraction of neutrons of first order,
 T_1 is the first order energy transmission,
 f_2 is the fraction of neutrons of second order,
 T_2 is the second order energy transmission,

and where reflections above second order are omitted. A constraint condition that Eq. (4.2a) is subject to is that $f_1 + f_2 = 1.0$.

For the Be(12 $\bar{3}$ 1) reflections the first and second order fractions were determined by Haas and Shore³² and they found $f_1 = 0.994$ and $f_2 = 0.006$.

Ordinarily T_2 is taken as 1.0 when no resonance appears at $4E$. In tungsten, however, a large resonance appears at 18.8 ev so that special care had to be given to the value of T_2 used in this study. In this case what was done was to measure T_2 over an energy range centered about $4E_0$. These measurements gave values of $T_2 = 0.98$ at an energy of 16.2ev and a value of $T_2 = 0.88$ at an energy of 17.0 ev. When a transmission measurement was performed at a nominal energy, E , the corresponding T_2 value at $4E$ was obtained by an interpolation of the linear T_2 vs. E plot.

From a consideration of the above the observed transmission values were corrected for second order contamination by the formula

$$T_1 = \frac{1}{0.994} (T_{obs} - 0.006T_2) \quad (4.2b)$$

and this is the value used to calculate the cross section in Eq. (4.1a)

2. Presence of Non-target Nuclei in Sample

The transmission study made on the D₂O solution of Na₂WO₄ required an additional correction. In this study a separate quartz cell of identical thickness containing only D₂O was used for the

³²R. Haas and F.J. Shore, Rev. Sci. Instr., 30, 17 (1959).

open beam measurements. When the ratio of the transmission of the two cells is taken the effects of the sodium and the oxygen part of the tungstate radical must be considered as well as the small D₂O unbalance between them. When these effects were calculated they introduced less than a 0.5 percent correction in the transmission results.

3. Effects of other Resonances

In a total cross section measurement at energy E, the quantity that is observed is the summation of contributions of all resonances appearing in the target material. For nuclei in which resonances are narrow with respect to their spacings, it is evident that the interference between levels is small and multilevel interference formulae are not necessary. For cases of this type the observed total cross section is well approximated by the relation

$$\sigma_t(E) = \sum_i [\sigma_{m,i}(E) + \sigma_{m,m}(E)] \quad (4.3a)$$

where σ_t is the observed total cross section,

$\sigma_{m,i}$ is the single-level Breit-Wigner partial absorption cross section given in Eq. (2.1a),

$\sigma_{m,m}$ is the single-level Breit-Wigner partial scattering cross section given in Eq. (2.2a), and

i is the index of summation over all resonances.

In the partial scattering cross section, $\sigma_{m,m}$, the interference between resonance and potential scattering is taken into account.

In an accurate resonance study the data required for analysis should include only the cross section of the single level under

investigation. Since, however, the measured value includes contributions from all resonances, a correction must be applied to the observed total cross section to take account of the effects of other resonances.

One of the difficulties in determining the effects of other resonances is the lack of information concerning the nuclear radius, R . Formulae, based on experimental measurements, have been offered that express R in terms of mass numbers. These formulae, however, were not considered sufficiently accurate for our purposes. Therefore a special measurement was made at an energy of 1.75 ev where the effects of the 4.14 ev resonance were negligible. At this energy the observed cross section was found to be 7.52 ± 0.13 barns. Application of this result to the evaluation of R is considered in the next paragraph.

A convenient expression for the total cross section can be obtained from Eq.(4.3a) as

$$\sigma_t(E) = 4\pi R^2 + \sum_l \frac{(0.651 \times 10^9) g \Gamma_m^0}{(E-E_0)^2 + \frac{\Gamma^2}{4}} \left[\frac{\Gamma_\gamma}{\sqrt{E}} + \Gamma_m^0 + 0.878 \times 10^{-3} R(E-E_0) \right] \quad (4.3b)$$

where $E, E_0, \Gamma, \Gamma_\gamma, \Gamma_m^0$ (the reduced neutron width at $\text{lev} = \frac{\Gamma_m^0}{\sqrt{E_0}}$) are in ev and R is in units of 10^{-13} cm. Here the first term in the summation is for capture, the second term is for resonance scattering, and the third term is interference scattering. For s-wave neutrons the potential cross section is given by

$$\sigma_p = 4\pi R^2 \quad (4.3c)$$

which appears in Eq.(4.3b). For tungsten nuclei where the levels are

well spaced the parameters of all significant resonances are available¹ as well as a listing of the isotopic abundances. Using these values on the right side of Eq. (4.3b) and the experimental result for σ_x at 1.75 ev on the left side, the equation reduces to a quadratic form in R which is immediately solvable. The evaluation of the quadratic equation gives as its positive root the value of

$$R = 9.92 \times 10^{-13} \text{ cm},$$

from which the potential cross section is evaluated to be

$$\sigma_p = 12.36 \text{ barns.}$$

The reason the measured cross section is less than the potential cross section is due to the negative contributions of the interference scattering terms, i.e. for $E = 1.75$ ev, the quantity $E - E_0$ for all tungsten resonances is negative.

The correction to the 4.14 ev resonance cross section attributable to other tungsten levels can be determined now. Re-using Eq. (4.3b) σ_x was calculated at several energies about the 4.14 ev resonance where all other resonance contributions were included. The calculations showed that σ_x varied little across the resonance with an average value of -6.4 barns. The correction due to other resonances was accomplished by adding 6.4 barns to all observed resonance cross sections.

C. Qualitative Features of the Data:

Before delving into the analysis of the data according to the various theories available, it is first desired to discuss qualitatively the general features of the resonance data which are summarized in Figures 5 and 6.

¹Hughes, op. cit.

1. Metallic Foil

In Figure 5 the resonance shapes shown are experimental curves measured at four different temperatures. The data for these curves have been corrected for the non-resonance effects previously considered in this chapter. In this figure the significant effect that temperature has on the observed peak cross section is readily observable as well as the change in the level's width. One surprising result was the relatively small difference between the 77.8°K curve and the 4.2°K curve as compared to the differences between the other curves. Lamb has indicated, however, that this comparison should be made in terms of the effective temperature of the sample in place of its real temperature. On this basis the effective temperatures are 130°K and 116°K , respectively, so that the difference noted is not as striking.

In Figure 5 there is observable an increase in the asymmetry of the curves as the temperature is lowered. Additionally there can be seen a shift in the center line of the resonance to higher energies as the temperature is increased. On physical grounds one understands this shift as being attributable to the neutron giving up a greater fraction of its incident energy to the recoil of the compound nucleus as the strength of the lattice binding is weakened. Therefore with less energy available for the nuclear process more energetic incident neutrons are required to excite the resonance.

2. Liquid Solution of Na_2WO_4

The experimental resonance shapes for the metal and liquid samples at 296°K are shown in Figure 6. Here small but real differences can be seen in the resonance shapes, especially on the high energy side of the resonance. Such differences have not been reported previously. The importance of this effect will be discussed in Chapter VI.

Fig. 5. Experimental resonance shapes in tungsten metal. The experimental points were omitted for clarity.

Fig. 6. Resonances shapes for metal target and liquid sample at 296° K. The two samples were studied simultaneously. The statistical uncertainty for all experimental points are well within the circles or triangles shown. For reproduction reasons it was found necessary to enlarge the sizes of the circles and triangles for the experimental points to approximately twice the calculated statistical uncertainties. The differences between the experimental data are more significant than indicated here.

CHAPTER V

ANALYSIS FOR THE PARAMETERS OF THE TUNGSTEN RESONANCE

In Chapter IV the experimental cross section data have been corrected for all non-resonance effects. Before the results of Doppler theories can be compared with experimental data it is first necessary to determine the resonance parameters. The objective of this chapter is the evaluation of the Breit-Wigner one-level parameters for the tungsten resonance.

A. The Complexities of Neutron Resonance Analysis:

1. Statement of the Problem

In general the pure Breit-Wigner shape for total cross section cannot be observed directly because of the effects of Doppler broadening. Moreover even the Breit-Wigner Doppler broadened resonance shape cannot be observed due to the finite resolving power of the spectrometer. The cross section curve observed in a resonance study is the result of two convolutions made on the Breit-Wigner shape by Doppler and resolution broadening.

The problem of proceeding from an observed resonance shape to its level parameters is extremely complex. The main reason for this complexity is that it is not possible to mathematically separate the Doppler and resolution effects so that the corrections for each cannot be made independently. It is inherent in the nature of the functions convoluted with the Breit-Wigner formulae that makes the reverse operation lack sensitivity with respect to the level parameters. Because of this insensitivity there have always been lingering doubts as to the uniqueness of reported parameters and their meaningfulness.

In the past many resonances were analyzed for their parameters using desk calculators, tables of Ψ -functions, special charts, etc. Due to the great amount of work involved it became necessary to introduce some practical approximations into the analysis. When these methods have been used the parameters obtained have usually been reported as being "satisfactory". The word "satisfactory" as it has been used has a spectrum of meanings ranging all the way from being crudely satisfactory to where the results are highly satisfactory. A good reliability test for the accuracy of resonance parameters is if they reproduced the observed cross section curve when substituted back into the cross section formulae. This reliability test, unfortunately, has been employed too infrequently.

2. Practical Formula for the Doppler Broadened Cross Section

In the case where the assembly of target atoms is assumed to behave as a free gas the Doppler broadened cross section was shown to be given by Eq. (2.6). This equation is awkward to use in analysis and a simplified form has been used instead. In the simplified form the effects of interference scattering are neglected and the resonance scattering term is multiplied by $\sqrt{\frac{E_0}{E}}$. The latter approximation introduces a very small error since over the energy range of a narrow resonance this term is very close to unity. Using these approximations Eq. (2.6) reduces to

$$\sigma_{MT}(\Delta, E) - \sigma_p \approx \sqrt{\frac{E_0}{E}} \sigma_0 \Psi(x, t) \quad (5.1a)$$

where the peak cross section, σ_0 , is given by

$$\sigma_0 = \sigma_{m, \gamma_0} + \sigma_{m, m_0} \quad (5.1b)$$

Eq. (5.1a) is the form of the Breit-Wigner Doppler broadened equation that has been used most frequently in resonance analysis. When the target is a solid absorber a rule that has been applied quite generally is to replace the sample's real temperature with its effective temperature in evaluating the Doppler width.

B. Discussion on Methods of Analysis:

1. Methods Available

The method that was used to determine the one-level parameters for the tungsten resonance is considered in the next section of this chapter. Before considering it, however, it is well to briefly describe methods of analysis which have been typical of work performed in the past. This is done to point out the inadequacy of such analysis techniques as applied to our particular study. Since we seek to explore microscopic details of the shape of one resonance it is necessary that our analysis be as rigorous and as objective as possible avoiding all approximations.

In the energy range below about 10 ev resonance analysis has generally been performed by either a "shape" method or an "area" method. The shape method is essentially a comparison between generated curves and the observed data. The generated curves, based on trial parameters, take due account for the effects of Doppler (usually according to Eq. (5.1a)) and resolution broadening. The parameters that yield the best fit between the theoretical curve and the experimental data were assigned to the level. The method is laborious so that it has been used for only a limited number of cases³³, however, many different approaches to the shape method have been made. Landon, Sailor and Wood³⁴

³³V.L. Sailor, Phys. Rev., 91, 53 (1953).

³⁴R.E. Wood, H.H. Landon and V.L. Sailor, Phys. Rev., 98, 639 (1955); H.H. Landon and V.L. Sailor, Phys. Rev., 98, 1267 (1955).

developed a simplified shape method that has been applied frequently.

Two basic requirements for the use of the shape method are that the resolution of the spectrometer be sufficiently "good" to observe the details of the resonance shape and that the resolution function be accurately known. The resolution is considered to be good when the resolution width is no larger than the natural width of the level. Where possible this method is to be preferred to the area method because it makes more sensitive use of the data.

For spectrometers with poorer resolution a less precise method for analysis has been developed. The area method³⁵ is ideally carried out on transmission curves made with two samples: a "thin" sample and a "thick" sample. The method is based on the areas above the transmission dips and these areas can be shown to be independent of the resolution function. Frequently it is not possible to satisfy the "thinness" condition for a sample so that samples of intermediate thickness have to be used. For samples of intermediate thickness the analysis is complicated and requires the use of special curves to perform successive approximations³⁶.

When the resolution of a spectrometer is poor the area method of analysis has to be used.

C. Analysis of the Tungsten Resonance:

1. Breit-Wigner Parameters for Tungsten

In the analysis of the observed tungsten resonance data, shown

³⁵E. Melkonian, W.W. Havens and L.J. Rainwater, Phys. Rev., 92, 702 (1953).

³⁶F.G.P. Seidl, D.J. Hughes, H. Palevsky, J.S. Levin, W.Y. Kato and N.G. Sjostrand, Phys. Rev., 95, 476 (1954); D.J. Hughes, J. Nuclear Energy, 1, 237 (1955).

in Figures 5 and 6, the shape method was used. At the resonant energy the resolution width was approximately two-thirds of the natural width of the level. The resolution width was known to be this because the resolution properties of the crystal spectrometer used in this study have been investigated thoroughly⁷.

It was possible to carry out a shape analysis of the tungsten resonance because of the availability of a modern high speed computer³⁷. By using a computer it is not necessary to make the simplifying approximations given by Eq. (5.1a).

Two separate studies for the parameters of the tungsten level have been reported previously^{38,39}, but they were judged to be insufficiently accurate for our purposes and a re-analysis was deemed necessary.

The evaluation of the nuclear radius, R , and the potential cross section, σ_p , were discussed in Section B of Chapter IV. These results were used throughout this study.

In the past the resonance energy was frequently determined by fitting a straight line through the center of the resonance and extending it to intersect the energy (abscissa) axis. This observed energy is not the true resonance energy, E_0 , but is actually greater by a small amount due to recoil. The true resonance energy is very difficult to determine because of the uncertainty in the amount of energy the incident neutron gives to the compound nucleus in recoil. In the case of an ideal gas or

⁷Sailor et al., op. cit.

³⁷The analysis of the tungsten resonance reported here was performed on the IBM 7094 computer located at Brookhaven National Laboratory.

³⁸H.H. Landon, Phys. Rev., 100, 1414 (1955).

³⁹J.R. Waters, J.E. Evans, B.B. Kinsey and G.H. Williams. Nuclear Physics, 12, 563 (1959).

an absorber that satisfies the effective temperature model requirements, the true Breit-Wigner E_0 value is less than this observed value by the free atom recoil energy. In general since target materials are rarely ideal the difference will be less than the free atom recoil energy but it is difficult to know precisely how much less. Standard practice is to quote the observed value for the resonance energy.

In the tungsten study the quantity labelled E_0 was determined as that value which gave the best fit of a theoretical curve to the experimental data neglecting recoil considerations entirely. In effect this amounted to assuming that the target nucleus is at rest in the laboratory coordinate system and possesses an infinite mass. The free atom recoil energy for W^{182} is about 0.0226 ev and the true resonance energy is approximately the value listed as E_0 less this amount.

In the analysis of the tungsten resonance the Doppler effect was accounted for by use of the effective temperature model. The required condition for the application of this model, Eq. (2.7f), is best satisfied when the sample is at the highest possible temperature where the Doppler width is largest and the chemical binding weakest. Because of this effective temperature model requirement the shape analysis to determine the radiation width, Γ_γ , and the scattering width, Γ_m , was carried out using the 825°K data. The parameters reported in this study for Γ_γ and Γ_m are based exclusively on the resonance shape at 825°K and are not the values which best fit all the observed resonance shapes at the different temperatures.

At the sample temperature of 825°K the effective temperature was calculated by Eq. (2.7d) to be 830°K. The Debye temperature, treated

as a constant in this study, was $310^{0.40}\text{K}$.

In order to explain how the shape fitting analysis was made; a brief outline of the calculations performed by the computer program is given in the next paragraph. No details of numerical methods or programming techniques are explained.

The Fortran program was compiled to do the following calculations.

1. For a set of trial parameters E_0 , Γ_γ and Γ_m , the Breit-Wigner Doppler broadened cross section was calculated according to Eq. (2.6) at each of the 27 experimental data points recorded at $825^{0.40}\text{K}$.
2. The results of step (1) were next converted to Doppler broadened transmission values using Eq. (4.1a).
3. At each energy the results of step (2) were then convoluted with the resolution function of the spectrometer according to Eq. (2.13).
4. The Doppler and resolution broadened transmission values of step (3) were then converted to cross section values by again using Eq. (4.1a).
5. At each experimental point the square of the difference between the theoretical cross section result of step (4) and the experimental cross section value was calculated.
6. These squared differences were then summed for the 27 points. This summation value (the residual) was taken to be a measure of the "goodness" of a theoretical fit to the experimental data. That combination of E_0 , Γ_γ and Γ_m which made the residual minimum was assigned to the level.

⁴⁰M.W.Holm, AEC Research and Development Report, IDO-16399 (1957).

In order to systematically arrive at the best set of parameters the following procedure was follows.

1. Using as initial values those results that had been reported in the literature for Γ_γ and Γ_m , E_0 was allowed to assume a set of values equally spaced. For a first round of calculations this grid of energy values was widely spaced. For each trial energy the residual was calculated.
2. A graph of Residual (barns²) vs. Energy was plotted. This graph had a parabolic shape from which one could estimate a first approximation value for E_0 that yielded a minimum residual.
3. Using this trial value for E_0 , both Γ_γ and Γ_m were next varied. Here Γ_γ was permitted to assume a set of values equally and widely spaced and so was Γ_m . In pairs of Γ_γ and Γ_m values the residual was calculated and the pair that had the new minimum residual was temporarily assigned to the level and superseded the original values.
4. To demonstrate a minimum in the residual when both Γ_γ and Γ_m were varied a two-dimensional "contour" map was drawn. The ordinate of this figure was Γ_γ and the abscissa axis was Γ_m . At each coordinate point where a residual value had been calculated, that value was assigned to the point. When lines of equal residual are drawn the figure takes on the appearance of a contour map very similar to an elevation map for a deep uneven hole. Such a drawing is shown in Figure 7 for the tungsten analysis.

Fig. 7. Isometric lines for the shape fitting procedure. The contour lines shown in the figure have been drawn as smooth lines and are in units of barns-squared. The resonance energy used for these calculations was 4.1422 eV. The minimum residual occurred for a capture width $\Gamma_\gamma = 0.0555$ eV and a scattering width $\Gamma_n = 0.000396$ eV. The scattering width here is uncorrected for isotopic abundance.

5. At this point in the analysis first approximation

values for E_0 , Γ_γ and Γ_m have been evaluated. To obtain more accurate results steps (1) to (4) were repeated with smaller grid ranges for E_0 and for $(\Gamma_\gamma, \Gamma_m)$ pairs until the final minimum residual was obtained.

The parameters obtained by the procedure outlined above for the tungsten resonance are given in Table I.

The theoretical shape generated with the parameters listed in Table I at a sample temperature of 825°K appears as curve C in Figure 8. The agreement between curve C and the experimental points shown in this figure is really quite good, but not perfect. When examined closely on a drawing to a much larger scale the details of the shape fit are more distinctly seen and small consistent differences are definitely present. It would appear that these differences are an indication that the condition for the effective temperature model, Eq. (2.7f), is not well satisfied at the temperature of 825°K.

In Figure 8 and in many of the following figures there appears at the bottom a triangular resolution function. Actually, however, the resolution function for the crystal spectrometer is a gaussian shape and it was the gaussian resolution function that was used in the analysis. In these figures the triangle gives only the width of the resolution function and the ordinate value is drawn to an arbitrary scale.

D. Comparison of Parameters:

1. Comparison with Previously Reported Results

Two previous measurements for the parameters of the tungsten

Table I. The Breit-Wigner parameters listed below are the best set of values that give agreement with the 825°K data. The analysis at this temperature is considered more valid than the analysis at other temperatures because of the Doppler effect. The Doppler effect was accounted for by use of Lamb's effective temperature model. The errors listed come from the analysis alone^a.

$$\begin{aligned}
 E_0 &= 4.1422 \pm 0.0005 \text{ ev}^{b,c} \\
 \Gamma_g &= 55.5 \pm 0.5 \text{ mev}^d \\
 \Gamma_m &= 1.50 \pm 0.01 \text{ mev}^e \\
 \sigma_0 &= 4502.3 \pm 50.5 \text{ barns}^f \\
 \sigma_p &= 12.36 \pm 2.10 \text{ barns} \\
 R &= (9.92 \pm 1.20) \times 10^{-13} \text{ cm} \\
 \mathcal{Q} &= 4.44 \pm 0.53 \text{ barn-ev}
 \end{aligned}$$

^aThe errors listed have to be increased somewhat to take care of systematic uncertainties, e.g. isotopic uncertainties, sample thickness, etc. The sample thickness was known to better than 1%.

^bThere is an inherent error in the absolute Bragg angle setting of the spectrometer which amounts to $\pm \frac{1}{4}$ minute of arc. This gives an energy uncertainty of ± 0.0058 ev.

^cThis value for E_0 is the best fitting value for the minimum residual as described in the text. The Breit-Wigner value is approximately 0.0226 ev less than this, i.e. the amount of the free atom recoil energy.

^d $\text{mev} = 10^{-3} \text{ ev}$

^e Γ_m is corrected for isotopic abundance.

^f σ_0 is not corrected for isotopic abundance.

Fig. 8. The total cross section of tungsten metal as a function of energy at 825° K. Curve A is the theoretical Breit-Wigner shape calculated for resonance parameters listed in Table I. Curve B results from operating on A with Doppler broadening according to the effective temperature model. Curve C was obtained by operating on B with the resolution function of the spectrometer. The cross section uncertainties are everywhere smaller than the diameter of the plotted points.

level have been reported^{38,39}. Table II summarizes the results obtained in this study (taken from Table I) and compares them with the other values reported.

In general sets of parameters derived from independent measurements and analyzed by different methods almost invariably show discrepancies outside the experimental errors. The comparison of our results with the values reported by Landon indicates disagreements in the parameters Γ_γ and Γ_m . Both sets of experimental data were recorded using the same crystal spectrometer at Brookhaven. In the work of Landon the analysis procedure was judged to be inferior in quality to that presently offered.⁴¹

In Table II the agreement between the parameters reported here are in better agreement with those of Waters et al. than they are with the results of Landon. In the comparison of results with Waters et al. one notices a slight disagreement in the value for Γ_γ and it is difficult to comment on why this disagreement exists. On this point it is worth mentioning that the analysis method of Waters et al. is very indirect and involves many approximations. It would be difficult to imagine how the accuracy of their method can be compared to the directness, simplicity and rigor of the shape method as it was applied here. One further observation is that our value for Γ_γ as reported in Table I is in better agreement with the constancy

³⁸Landon, op. cit.

³⁹Waters et al., op. cit.

⁴¹H.H. Landon, private communication

TABLE II
COMPARISON OF BREIT-WIGNER PARAMETERS
FOR TUNGSTEN (W^{182}) RESONANCE

	Reported in This Paper	Previously Reported	
		Landon ^a	Waters et al. ^b
Sample Material	Metal	Metal	Metal
Neutron Source	Reactor	Reactor	15 Mev electron linear accelerator
Experimental Studies	Neutron Transmission	Neutron Transmission	Neutron Transmission, Elastic Scattering, and Capture γ -rays
Method of Analysis	Shape	Modified Shape	Modified Area plus others
Sample Temperature ($^{\circ}K$)	825	296	296
E_0 (ev)	4.1422	4.14	4.14
Γ_{γ} (mev) ^c	55.5 ± 0.5	46 ± 2	49.3 ± 4.7
Γ_m (mev)	1.50 ± 0.01	1.43 ± 0.03	1.48 ± 0.06

^aSee reference 38.

^bSee reference 39.

^cmev = $10^{-3}EV$

of radiation widths for other W^{182} levels than is the value of Waters et al.

2. Comparison with Resonance Shapes at 296°K

The parameters determined in this study for the tungsten resonance were based on measurements where the sample was kept at 825°K . The parameters reported by Landon and by Waters et al. were based on measurements where the targets were kept at 296°K . In order to test whether this difference in temperature could account partially for the discrepancy in the reported parameters it was decided to extend our analysis to include the resonance shapes measured at 296°K . These curves are shown in Figure 6. Of course it is well known that resonance parameters are not temperature dependent, but as a matter of general interest these calculations were performed.

At room temperature the resonance shape was measured for two different samples: a metal target and a heavy water solution of Na_2WO_4 . The parameters that were obtained for both of these shapes are listed in Table III. In this analysis the Doppler effect was accounted for by using the effective temperature model for the metal and the gas model for the liquid.

The parameters in Table III reveal that the values for Γ_{γ} and Γ_m for both the metal and the liquid sample at room temperature are in satisfactory agreement with the values reported by Waters et al. This agreement, however, is not to be interpreted as having these values supersede the results given in Table I. The reasons for this are: (1) the required condition for the application of the effective temperature model, Eq. (2.7f), is less well satisfied for the metal

Table III. Sets of Breit-Wigner parameters that were obtained by applying the shape fitting procedure described in the text to three different experimental shapes. Analysis for the metal sample was done in terms of the effective temperature model. For the heavy water solution of Na_2WO_4 , the ideal gas model was applied.

Sample Material	Metal	Metal	Solution of $\text{D}_2\text{O} - \text{Na}_2\text{WO}_4$
Doppler Broadening Model	Effective Temperature	Effective Temperature	Ideal Gas
Sample Temperature ($^{\circ}\text{K}$)	825	296	296
Effective Temperature ($^{\circ}\text{K}$)	830	312	
E_0 (ev)	4.1422	4.1413	4.1404
Γ_{γ} (mev) ^a	55.5	53.2	53.0
Γ_m (mev)	1.50	1.48	1.47

$$^a_{\text{mev}} = 10^{-3} \text{ ev}$$

at 296°K than it is for the metal at 825°K, and (2) the gas model used to explain the Doppler effect for the liquid solution cannot be theoretically justified.

The interpretations given in the last paragraph find added support when theoretical shapes based on the parameters listed in Table III are compared with the experimental data. For the liquid the calculated shape is shown as curve C in Figure 9. A similar figure for the metal was prepared but it has been omitted here. An examination of these figures reveals that the theoretical curves fit the experimental data rather poorly. A comparison between these two figures indicates that the agreement is poorer for the liquid than for the metal. Each of the room temperature fits is in poorer agreement with the corresponding data than are the 825°K results appearing in Figure 8. For the metal sample this increased disagreement supports the theoretical interpretation that the effective temperature model becomes increasingly poorer to describe the Doppler effect as the temperature of the sample is lowered.

For the data recorded at 296°K the theoretical interpretation of the metal data is in better agreement than the theoretical interpretation for the liquid data. This conclusion is an indication that the effective temperature model accounts for the Doppler effect in a metal more accurately than does the gas model for the liquid.

E. Resonance Shapes Compared With Experimental Data at Different Temperatures:

The resonance parameters Γ_γ and Γ_m are nuclear properties which are not temperature dependent. In the center of mass coordinates the resonance energy, E_0 , is also temperature independent, but because

Fig. 9. The total cross section of tungsten as a function of energy at 296°K . The sample was a heavy water solution of sodium tungstate. The resonance energy of 4.1404 eV was obtained by the shape fitting procedure described in the text. Curve A is the theoretical Breit-Wigner shape calculated for resonance parameters listed in Table III. Curve B results from operating on A with Doppler broadening according to the gas model. Curve C was obtained by operating on B with the resolution function of the spectrometer. The cross section uncertainties are everywhere smaller than the diameters of the plotted points.

of recoil effects it is very difficult to know this value to high accuracy. To compare the experimental results on tungsten metal at temperatures of 296°K, 77.8°K and 4.2°K with the theoretical predictions of Lamb's effective temperature model at these sample temperatures more analysis was undertaken.

In the additional analysis the radiation width and the scattering width as reported in Table I were held fixed. The shape fitting procedure was then performed to determine the best value for the resonance energy that would yield the minimum residual. These results are listed in Table IV.

In Table IV one notices that the resonance energy monotonically increased as the temperature increased. This change in the apparent resonance energy between the 4.2°K data and the 825°K data is believed to be real and it is equivalent to 17 percent of the free atom recoil energy. On physical grounds one understands this increase as being attributable to the neutron giving up a greater fraction of its incident energy to the recoil of the compound nucleus as the strength of the lattice binding is weakened. Therefore with less energy available for the nuclear process more energetic incident neutrons are required to excite the resonance.

Figures 10, 11, and 12 show the total cross section of tungsten metal as a function of energy at temperatures of 296°K, 77.8°K and 4.2°K, respectively. In these figures the curves labelled C are theoretical based on the effective temperature model and the open circles are the experimental data points corrected for the non-resonance effects discussed in Chapter IV.

Table IV. Independent determinations of the apparent resonance energy. The values for E_0 listed below were obtained as the best fits of theoretical curves to the experimental data recorded at different temperatures. These values are in error from the true E_0 by approximately the free atom recoil energy^a. The radiation width, $\Gamma_r = 0.0555$ ev, and the scattering width, $\Gamma_m = 0.00150$ ev, were held fixed in these calculations.

Sample Temperature (°K)	Effective Temperature ^b (°K)	Best Fit for Resonance Energy (°K)
825	830	4.1422 ev
296	312	4.1413 ev
77.8	130	4.1400 ev
4.2	116	4.1384 ev.

^aThe free atom recoil energy for W^{182} is approximately 0.0226 ev.

^bThe Debye temperature for tungsten is 310°K.

Fig. 10. Total cross section of tungsten metal as a function of energy at 296° K. The resonance energy of 4.1413 eV was obtained by the shape fitting procedure described in the text. The radiation and scattering widths used here are reported in Table I. Curve A is the theoretical Breit-Wigner shape. Curve B results from operating on A with Doppler broadening according to the effective temperature model. Curve C is obtained by operating on B with the resolution function of the spectrometer. The cross section uncertainties are everywhere smaller than the diameter of the plotted points.

Fig. 11. Total cross section of tungsten metal as a function of energy at 77.8° K. The resonance energy of 4.1400 eV was obtained by the shape fitting procedure described in the text. The radiation and scattering widths used here are reported in Table I. Curve A is the theoretical Breit-Wigner shape. Curve B results from operating on A with Doppler broadening according to the effective temperature model. Curve C is obtained by operating on B with the resolution function of the spectrometer. The cross section uncertainties are everywhere smaller than the diameter of the plotted points.

Fig. 12. Total cross section of tungsten metal as a function of energy at 4.2° K. The resonance energy of 4.1384 eV was obtained by the shape fitting procedure described in the text. The radiation and scattering widths used here are reported in Table I. Curve A is the theoretical Breit-Wigner shape. Curve B results from operating on A with Doppler broadening according to the effective temperature model. Curve C is obtained by operating on B with the resolution function of the spectrometer. The cross section uncertainties are everywhere smaller than the diameter of the plotted points.

CHAPTER VI

COMPARISON OF DOPPLER THEORIES WITH EXPERIMENTAL DATA

Let us now use the Breit-Wigner parameters which have been derived (Chapter V, Table I) to calculate the Doppler broadened resonance shape. Shapes have been calculated using the theories of Lamb, Nelkin - Parks, and Egelstaff - Holt. Comparisons of these shapes with the experimental data are presented in this chapter.

A. Additional Corrections to Data:

1. Resolution Correction

The observed tungsten data appearing in Figures 8 to 11 include the effects of Doppler and resolution broadening. In order to compare calculated Doppler broadened resonance shapes with experimental results a resolution correction must be applied to the data. Since it is not possible to analytically separate the Doppler and resolution effects a numerical method must be used.

The resolution correction for the 825⁰K data was determined by a simple but accurate method. Using the parameters given in Table I the computer calculations gave separate results for the resonance shape Doppler broadened and Doppler plus resolution broadened. From these calculations a resolution correction factor was obtained at each experimental energy by taking the ratio of the Doppler broadened value to the corresponding Doppler plus resolution broadened value. The experimental cross section data were corrected for the effects of resolution by being multiplied by these factors.

The resolution corrections applied at the other temperatures were evaluated in a similar manner but with one essential modification.

At each temperature "artificial" resonance parameters were determined that yielded the best fit of a calculated curve to the experimental data. These parameters, listed in Table V, were used to determine the resolution correction factors for the remaining experimental data. It is important to emphasize that these parameters are artificial and were used only for the purpose of making the resolution correction more accurate.

Resolution correction curves for tungsten metal at four different temperatures are given in Figure 13. At a given energy and for a given temperature the ordinate value here is the resolution correction factor. Multiplication of experimental cross section by this ratio yielded the total cross section corrected for resolution distortion. At the resonance peak the correction became more severe as the temperature was reduced varying from 3.1 percent at 825°K to 13.2 percent at 4.2°K.

The resolution corrections made on the Na₂WO₄ liquid data were performed using the parameters in Table III. The resolution correction curve for this sample is given in Figure 14. The peak resolution correction here was 7.0 percent.

2. Corrections for Potential and Resonance Scattering

The Doppler theories which we are examining have made the assumptions that the resonance under consideration is a pure capture level, in particular, the effects of the interference term in the resonance scattering have been neglected. It should be noted that all resonances possess finite scattering since the width Γ_m is

Table V. Independent sets of Breit-Wigner "parameters"

for the tungsten resonance. At each temperature they represent the best combinations that fit the experimental data. The Doppler effect was accounted for in terms of the effective temperature model. The parameters obtained at the sample temperature of 825°K are considered the true Breit-Wigner parameters. The other parameters are artificial and were obtained only to determine the resolutions corrections to the data at the different temperatures.

Sample Temperature (°K)	825 ^a	296 ^b	77.8	4.2
Effective Temperature (°K)	830	312	130	110
E ₀ (ev)	4.1422	4.1413	4.1400	4.1384
Γ _γ (mev) ^c	55.5	53.2	52.2	52.2
Γ _m (mev)	1.50	1.48	1.47	1.47

^a The parameters at 825°K were listed previously in Table I.

^b The parameters at 296°K were listed previously in Table III.

^c mev = 10⁻³ ev

Fig. 13. The resolution correction curves for tungsten metal at different temperatures. The minimum peak correction was 3.1 percent at 825°K and the maximum peak correction was 13.2 percent at 4.2°K.

Fig. 14. The resolution correction curve for the D_2O - Na_2WO_4 solution sample at 296°K . The peak correction was 7.0 percent

never zero. Therefore in order to compare our data with these theories it is necessary that we subtract out the scattering contribution to the total cross section. For this resonance in tungsten $\Gamma_s/\Gamma = 0.026$ so that the scattering cross section is only a small fraction of the total cross section, and hence the effect of the scattering cross section on the shape of the resonance is extremely small.

The contributions of all scattering cross sections to the observed total cross section, resolution corrected, were evaluated in the following way. Using the resonance parameters listed in Table I the Doppler broadened terms for resonance, interference and potential scattering were summed at each energy where cross sections were made and at each temperature. These values were then subtracted from the corresponding experimental data.

When this last correction had been applied, the experimental cross section data that remained was only the capture part of the total cross section. In all subsequent figures the calculations based on the theories of Lamb, Nelkin-Parks, and Egelstaff-Holt are compared with the observed Doppler broadened capture cross section only.

B. Previous Experimental Research on the Doppler Effect:

The experimental results of this dissertation are compared with the Doppler theories in the next three sections of this chapter. Before considering them, however, it is well to review briefly the results of several previous research efforts that were also directed towards studying the Doppler effect on neutron resonances.

The first direct experimental test of the theories of Bethe-Placzek and Lamb was made in 1953 by Landon⁴² in which the peak

⁴²H.H. Landon, Phys. Rev., 94, 1215 (1954).

cross section of the 1.26 ev resonance in rhodium was measured as a function of temperature. The analysis for this data was performed in terms of Eq. (5.1a) using both the real temperature of the sample (Bethe-Placzek) and the effective temperature of the sample (Lamb). Landon concluded that his results were in reasonable agreement with either of the two theories because at each temperature there was so little difference between the real and effective temperatures. If the resolution of the spectrometer had been better, it is possible that Landon would have been able to distinguish between these two results.⁴³

A more comprehensive Doppler study was reported in 1962 by Jackson and Lynn⁴⁴. In this work the line shapes of two resonances were measured at a series of different temperatures. Measurements were made on the 6.71 ev resonance in osmium metal at 297°K and 105°K, and on the 6.65 ev resonance in uranium metal at 297°K, 77°K and 4°K. In addition the uranium study was repeated but with the sample modified to uranium oxide, U₃O₈.

The general conclusions arrived at from this research were:

1) for the case of osmium the effective temperature model predicted results that were within the accuracy of the experiment at both temperatures, 2) for the uranium resonance using the metal target Lamb's model faithfully reproduced the data at 297°K, but at the lower temperatures

⁴³In 1954 the resolution of the crystal spectrometer was improved significantly by the addition of Soller collimators. A description of these improved resolution properties is given in reference 7. The improved resolution was used to record the data reported here.

⁴⁴H.E. Jackson and J.E. Lynn, Phys. Rev., 127, 461 (1962)

the results were inconsistent with this approximation, and 3) for U_3O_8 sample the discrepancies between Lamb's theory and the experimental data became even more pronounced at the lower temperatures.

Jackson and Lynn also calculated theoretical line shapes using the expansion equation of Nelkin and Parks. These calculations indicated that the higher terms of the expansion do not make significant changes over Lamb's results.

They also carried out an additional analysis by replacing the energy transferred to the lattice by the recoiling nucleus by an expansion expression. Limiting the crystal to a simple Einstein single-frequency model, the results they calculated for the uranium resonance at $4^{\circ}K$ are in better agreement with the data than are the results of the effective temperature model (a Debye frequency spectrum). For the $4^{\circ}K$ results in U_3O_8 a Nernst-Lindemann model, i.e. a double Einstein frequency model, was found to give the best agreement.

C. Calculations for the Theory of Lamb Compared with Experimental Data:

1. Resonance Shapes for Metal Sample

The calculations for the tungsten resonance based on the effective temperature model of Lamb are shown in Figures 15 to 18 inclusive. Figure 19 is a composite of these four drawings. The Breit-Wigner parameters used to generate these shapes are given in Table I; the best fitting resonance energies used at each temperature are listed in Table IV.

Figure 15 shows the calculated resonance shape as a solid line at $825^{\circ}K$. The best agreement between the calculations based on Lamb's theory and the observed data appears at the highest temperature. Just

Fig. 15. Lamb's effective temperature model results for tungsten metal at 825°K . The solid line was calculated using the resonance parameters listed in Table I. The experimental points have been corrected for resolution and neutron scattering effects.

Fig. 16. Lamb's effective temperature model results for tungsten metal at 296°K. The solid line was calculated using the resonance energy of 4.1413 eV and the other parameters listed in Table I. The experimental points have been corrected for resolution and neutron scattering effects.

Fig. 17. Lamb's effective temperature model results for tungsten metal at 77.8°K. The solid line was calculated using the resonance energy of 4.1400 eV and the other parameters listed in Table I. The experimental points have been corrected for resolution and neutron scattering effects.

Fig. 18. Lamb's effective temperature model results for tungsten metal at 4.2°K . The solid line was calculated using the resonance energy of 4.1384 eV and the other parameters listed in Table I. The experimental points have been corrected for resolution and neutron scattering effects.

SHAPE VARIATIONS WITH TEMPERATURE

Fig. 19. Shape variations with temperature according to Lamb's effective temperature model. This drawing is a composite for figures 15 through 18 inclusive.

from the nature of Lamb's theory it should be best at the highest temperature. The good agreement between theory and data shown in Figure 15 demonstrates that the parameters obtained from the analysis do indeed give a good fit and these parameters are to be preferred to those obtained at lower temperatures.

It should be noted, however, that even though the fit in Figure 15 is good, it is still not perfect. Different sections of the resonance indicate there are consistent deviations between the solid line and the experimental points. Although these differences appear to be quite small in the figure, they are significant departures and the departures get amplified as the temperature is lowered. At 825°K the ratio $\frac{\Gamma + \Delta}{2\theta}$ for the tungsten level is equal to 2.5 only. Undoubtedly the reason for the shape discrepancies is that this value is not large enough to allow the effective temperature model to be applied rigorously. The required condition for the application of this model is given in Eq. (2.7f).

As the sample temperature was lowered the application of the theory of Lamb to account for the Doppler effect becomes less adequate as can be seen in Figures 16, 17 and 18. Nevertheless, at all temperatures it does represent a good first order approximation to the resonance data.

The solid line in Figure 20 represents the theoretical Doppler broadened peak cross section as a function of temperature according to the theory of Lamb. In this figure one notices that the theory underestimates the peak value as the temperature of the sample is reduced. The inadequacy of the theory about the resonance peak is

Fig. 20. Peak cross section of the 4.14 ev tungsten (metal) resonance vs. temperature. The solid line was calculated using Lamb's effective temperature model and the experimental points were plotted at their effective temperatures. The data points have been corrected for resolution and neutron scattering effects.

particularly apparent in Figures 16, 17 and 18. The unevenness of the data about the peak suggests the possible presence of the fine structure predicted by Lamb in the case of intermediate binding. The resolution function of the spectrometer is, however, too wide to allow a more definite conclusion.

2.. Resonance Shape for Liquid Solution.

The best fitting parameters obtained with a sample consisting of a D_2O solution of sodium tungstate are given in Table III. These values were determined by treating the Doppler broadening effect in terms of the ideal gas model. The disagreement between the true parameters of Table I and those of Table III is believed to be attributable to the inadequacy of this approximation.

A calculated Doppler broadened resonance shape using the results of Table III appears in Figure 21 as a solid line. The agreement between this line and the data is poor. If a comparison were made of the results appearing in Figure 21 and those of Figure 16, it would be concluded that the effective temperature model more faithfully reproduces the data taken on a metal sample than does the ideal gas model for the liquid sample. In this particular instance it can be added that the metal data agreement, Figure 16, would have been better if its best fitting parameters (also given in Table III) were used in place of the true level parameters.

D. Calculations for the Theory of Nelkin and Parks Compared with Experimental Data:

The Nelkin and Parks theory was explained briefly in Section D of Chapter II. The practical result of this theory is that the Doppler broadened resonance shape is expressed as an expansion equation, Eq.(2.10a),

Fig. 21. Ideal gas model results for tungsten resonance. The sample was a heavy water solution of sodium tungstate at 296°K. The solid line was calculated using the tungsten parameters listed in Table III. The experimental points have been corrected for resolution and neutron scattering effects.

the leading term of which is Lamb's effective temperature model.

In order to examine the effectiveness of this theory Eq.(2.10a) was programmed for the computer at Brookhaven. Previously a program for this equation had been compiled independently at Argonne National Laboratory for use on the computer GEORGE. Both of these programs were used for the tungsten resonance and the separate computer calculations are in agreement.

The calculated resonance shapes appear as the dashed lines in Figure 22 to 25 inclusive. Figure 27 is a composite of these four drawings. The curves were generated using the Breit-Wigner parameters listed in Table I; at the different temperatures the best fitting resonance energies listed in Table IV were used.

Figure 22 compares the predicted resonance shapes for both the theories of Lamb and Nelkin-Parks at 825°K. In this figure the calculated resonance shape based on the Nelkin-Parks theory reproduces the resonance data more accurately than does the theory of Lamb. The small discrepancies in shape that were previously noted at this temperature in Figure 15 have now almost disappeared.

The Nelkin-Parks theory is applied most appropriately at the elevated temperatures where the recoil is greatest. At temperatures of 296°K, Figure 23, and 77.8°K, Figure 24, the results of this new theory appear to agree better with the data than does Lamb's results, but even for the new theory small discrepancies remain. Figure 25 shows the results when the sample was at 4.2°K. At this temperature both the results of Lamb and Nelkin-Parks are poor and it is meaningless to consider which is better.

Fig. 22. Resonance shape calculated according to the theory of Nelkin and Parks at 825°K. The dashed line was calculated using the resonance parameters listed in Table I.

Fig. 23. Resonance shape calculated according to the theory of Nelkin and Parks at 296°K. The dashed line was calculated using the resonance energy of 4.1413 eV and the other parameters listed in Table I.

Fig. 24. Resonance shape calculated according to the theory of Nelkin and Parks at 77.8°K. The dashed line was calculated using the resonance energy of 4.1400 eV and the other parameters listed in Table I.

Fig. 25. Resonance shape calculated according to the theory of Nelkin and Parks at 4.2°K. The dashed line was calculated using the resonance energy of 4.1384 eV and the other parameters listed in Table I.

NELKIN AND PARKS
EXPANSION TERMS
SAMPLE TEMPERATURE 4.2 °K

Fig. 26. Individual terms of the Nelkin and Parks expansion equation calculated for the sample temperature of 4.2°K. The effect of these terms on the resonance shape appear in Figure 25 as the dashed line; omitting these terms gives the solid line.

SHAPE VARIATIONS WITH TEMPERATURE

Fig. 27. Shape variations with temperature according to the theory of Nelkin and Parks. This drawing is a composite for Figures 22 through 25 inclusive.

The contributions made by the higher order terms of Eq. (2.10a) to the total cross section are of general interest in neutron resonance analysis. To demonstrate their individual contributions Figure 26 was prepared. In this figure the 4.2°K results are shown because at this temperature the contributions made by the individual terms are largest. If the results at 825°K were presented both the third and fourth terms would not appear because they are almost zero at this temperature.

In Figure 26 it is clear that the first and second terms dominate the contributions made to the total cross section. The contributions made by the fourth term are greater than the third term which indicates the series is not convergent.

The Doppler broadened peak cross sections according to the Nelkin-Parks theory is shown in Figure 28. In this figure one sees that the peak values are above those of Lamb's calculations. This is correct as can be seen in a particular instance by referring to Figure 26. Here the second term makes a greater positive contribution to the cross section than the negative value of the fourth term leaving a net positive increase in the peak cross section. The first and third terms do not make contributions.

E. Calculations for the Theory of Egelstaff and Holt Compared with Experimental Data:

In the preceding sections of this chapter it was shown that Doppler broadened calculations based on the formulae of Lamb and Nelkin-Parks have their most notable success at the highest sample temperatures. In both theories lattice vibrations are interpreted in

Fig. 28. Peak cross section of the 4.14 ev tungsten (metal) resonance vs. temperature. The dashed line was calculated according to the theory of Lamb and the solid line was calculated using the theory of Nelkin and Parks. The experimental points were plotted at their effective temperatures.

terms of the Debye model which is applied frequently to physical situations where the details of crystal vibrations are not important. At lower temperatures part of the discrepancy between the calculated resonance shapes and the data is undoubtedly due to the limitations imposed on these theories by not using more detailed frequency spectra. The extent to which the details of frequency spectra actually affect cross section calculations is not known clearly. An additional factor to be considered in practice is that the large resolution widths of spectrometers generally make the observed cross section data even less sensitive to these details.

The Egelstaff-Holt theory was discussed briefly in Chapter II. As indicated this theory does not limit the analysis to any particular frequency distribution. The computer program, LEAP, used to calculate the Scattering Law is capable of accepting almost any phonon spectrum that the user may wish to insert. Ideally one would use only those spectra which have been experimentally determined.

A preliminary attempt to use the Egelstaff-Holt method for the tungsten resonance has been made using a Debye spectrum. A Debye temperature of 310°K was used which is the same value that was used in the other calculations. The results for the Egelstaff-Holt calculations at 825°K , 296°K and 77.8°K are shown as dashed lines in Figures 29, 30 and 31, respectively⁴⁵. Figure 32 is a composite of

⁴⁵Further investigation at the sample temperature of 4.2°K is required. Egelstaff has suggested (private communication) that at this temperature a possibility exists that the rather large range of α and β values are preventing the calculations from running to completion.

Fig. 29. Resonance shape calculated according to the theory of Egelstaff and Holt at 825°K. The resonance parameters used are listed in Table I. The positioning of the resonance shape was determined visually.

Fig. 30. Resonance shape calculated according to the theory of Egelstaff and Holt at 296°K. The resonance parameters used are listed in Table I. The positioning of the resonance shape was determined visually.

Fig. 31. Resonance shape calculated according to the theory of Egelstaff and Holt at 77.8°K. The resonance parameters used are listed in Table I. The positioning of the resonance shape was determined visually.

SHAPE VARIATIONS WITH TEMPERATURE

Fig. 32. Shape variations with temperature according to the theory of Egelstaff and Holt. This drawing is a composite of Figures 29 through 31 inclusive.

these three drawings.

An inspection of the results at 825°K and 296°K reveals that the resonance shapes calculated with this theory are in very good agreement with the data. In these figures the solid lines represent the calculations based on Lamb's effective temperature model. Except for the peak region at 825°K the results of the Egelstaff-Holt calculations appear to be in better agreement with the data than are Lamb's.

The results of the new calculations at 77.8°K are shown in Figure 31. At this temperature the agreement between theory and data is poor and the reason for this disagreement is not understood at present.

CHAPTER VII

CONCLUSION

This thesis represents an intensive research effort to understand the effect of Doppler broadening on a neutron resonance. To date experimental tests of the theories have been limited, although to some extent all resonance analysis might be interpreted as tests of the gross features of Lamb's effective temperature theory. In view of the enormous research effort that has been spent on resonance studies it is clear that more rigorous experimental studies of Doppler broadening are appropriate. This dissertation is offered to partially fulfill this need for highly detailed precision data.

The neutron resonance in tungsten at 4.14 ev was chosen for study because of its particularly favorable characteristics: well isolated from other resonances, it has a narrow width, the scattering contribution is small, and since W^{182} is an even-even nucleus the resonant spin is known unambiguously. Another important factor is that the physical properties of tungsten permit its use over a wide range of temperature. In addition, the resonant energy lies within the range of good spectrometer resolution. This latter aspect is particularly important because it was necessary that the resonance shape should be observed in fine detail if the theories were to be tested to high accuracy. Once these theories have been tested to high precision in this energy range for one resonance, they can be applied with confidence to other resonances at energies where experimental limitations do not permit detailed observations of the shape.

The conclusions of this research are graphically summarized in Figures 15 to 32 given in Chapter VI. In these figures the Doppler

broadened resonance shapes calculated according to the theories of Lamb, Nelkin-Parks, and Egelstaff-Holt are compared with the experimental data. The general conclusions which follow from these comparisons are:

1. Lamb's effective temperature model provides a good first order fit to the experimental data at all sample temperatures. It should be noted that as the sample temperature increased the agreement between theory and data becomes better as indicated in Figures 15 to 18. However, a close examination of the results at 825°K (Figure 15) reveals that even at this high temperature Lamb's theory did not reproduce the experimental data perfectly. For this case $\frac{\Gamma + \Delta}{2\theta} = 2.6$ which apparently is insufficient to satisfy Lamb's model rigorously.
2. The calculations at 296°K show that the effective temperature model is superior to interpreting the Doppler effect for a metal target than the ideal gas model is for a liquid sample. This result indicates that studies directed to evaluating resonance parameters to high accuracy should use metal samples whenever possible.
3. The calculations based on the theory of Nelkin and Parks (Figures 22 to 24) are in better agreement with the data than are the results of Lamb's theory. It would appear that future investigations directed towards determining Breit-Wigner parameters of a resonance to high accuracy should use the Nelkin-Parks theory to account for the Doppler effect. It is surprising that the results of Jackson and Lynn on the 6.65 ev resonance in uranium did not reveal better agreement with the Nelkin-Parks theory. Since the uranium resonance is significantly narrower than the

tungsten resonance it should have distinguished more sensitively between the two theories. However, unless the experimental study is done to high accuracy, the difference could not have been detected.

4. The results reported here on the Egelstaff-Holt theory are only preliminary. The agreement between the calculated and observed resonance shapes at 825°K (Figure 29) and at 296°K (Figure 30) give evidence for the validity of this method. It would be interesting to perform additional calculations using several different phonon spectra in addition to the Debye spectrum. Results using an experimentally determined spectrum would be of special interest.
5. Future studies of Doppler broadening afford the possibility for gaining information about lattice dynamics. Such work would require data of even greater precision than reported here, and particularly better spectrometer resolution will be needed. It is doubtful whether this approach to the study of phonon spectra would compete with methods now under development which involve the inelastic scattering of cold neutrons. However, it appears likely that Doppler studies could supplement the inelastic scattering data by amplifying some features of the problem which cannot be adequately resolved by conventional techniques.

APPENDIX A *

Lamb begins his development by using a well-known formula from radiation theory which gives the transition probability between two states via an intermediate state as

$$W(B\{\beta_s\} \vec{k}; A\{\alpha_s\} \vec{p}) = \left| \sum_{\{m_s\}} \frac{\langle \Psi_{B\{\beta_s\}} | H^{(2)} | \Psi_{C\{m_s\}} \rangle \langle \Psi_{C\{m_s\}} | H^{(1)} | \Psi_{A\{\alpha_s\}} \vec{p} \rangle}{E_0 - E + E(\{m_s\}) - E(\{\alpha_s\}) + \frac{i\Gamma}{2}} \right|^2 \quad (A.1)$$

The essential fact used here is that the transition probability for a given transition is proportional to the square of the matrix element connecting the states involved. The formula is derived in first order, time dependent perturbation theory.

Under the assumption that the lattice forces are harmonic the curly bracket notation, $\{...\}$, denotes a state of the crystal by a set of numbers which give the phonon occupation numbers in the various normal modes of lattice vibration.

$H^{(1)}$ is the energy operator which described the interaction between the incident neutron and the target atom. $\Psi_{A\{\alpha_s\}} \vec{p}$ is the wave function for the initial state where the neutron with momentum $\vec{p}$ is incident on a nucleus A which in turn is held in a lattice of state $\{\alpha_s\}$. $\Psi_{C\{m_s\}}$ is the wave function for the

*The treatment given here is essentially a re-statement of Lamb's paper. It is felt that this development satisfied a need of verbally "filling-in" some items that are possibly too succinctly expressed in the original papers.

intermediate compound nucleus state held in a lattice state $\{m_s\}$. Therefore $H^{(1)}$ matrix element connects the initial state of the system to an intermediate state.

$H^{(2)}$ is the energy operator which describes the radiative decay of the compound nucleus with the emission of a gamma ray of wave vector $\vec{k}$. $\psi_{B\{\beta_s\}}(\vec{r})$ is the wave function which describes the final state where the residual nucleus B is in its ground state and the lattice is in state $\{\beta_s\}$. Therefore the second matrix element connects the intermediate state with the final state.

The summation is over all intermediate crystal states in which the nucleus is in the excited state C .

The recoil of the compound nucleus transfers energy to the lattice of the crystal causing a transition from the initial state with an energy $E(\{\alpha_s\})$ to the intermediate state with an energy $E(\{m_s\})$. Energy conservation is implicit here since the intermediate states with energy $E(\{m_s\})$ are only possible if

$$E(\{m_s\}) + E_0 = E(\{\alpha_s\}) + E. \quad (A.2)$$

This energy transferred to the lattice causes a change to take place in the phonon occupation numbers of the normal modes of vibration of the crystal, so that another way to express the recoil energy is

$$E(\{m_s\}) - E(\{\alpha_s\}) = \sum_s (m_s - \alpha_s) \hbar \omega_s. \quad (A.3)$$

To be perfectly accurate the value for the half width of the level, Γ , is equal to the nuclear width plus some dependence on the intermediate crystal states. The summation over intermediate states is greatly simplified, however, if the dependence of Γ on the state of the lattice is neglected which is what is done.

These matrix elements are too general for calculation so that it is necessary to change them to more explicit forms. To do this Lamb assumes that the nucleon motions inside the nucleus can be separated from the center of mass motion of the nucleus in the lattice so that the probability for formation of the compound nucleus is independent of the lattice effects. This separation of nucleus and lattice is reasonably good because the lattice forces are very weak compared to the strong internal nuclear forces so that the crystalline binding acts only on the center of mass motion of the nucleus and does not disturb the nucleon motion. Because of this separation the wave functions $\psi_{A\{\alpha_s\}} \vec{p}$, $\psi_{C\{m_s\}}$, and $\psi_{B\{\beta_s\}} \vec{k}$ can be expressed as products of separate nuclear and lattice wave functions with the following replacements being made:

$$\langle \psi_{C\{m_s\}} | H^{(1)} | \psi_{A\{\alpha_s\}} \vec{p} \rangle \longrightarrow M_C \langle \psi_{\{m_s\}} | e^{i \vec{p} \cdot \vec{X}_L / \hbar} | \psi_{\{\alpha_s\}} \rangle \quad (A.4)$$

and

$$\langle \psi_{B\{\beta_s\}} \vec{k} | H^{(a)} | \psi_{C\{m_s\}} \rangle \longrightarrow M_{rad} \langle \psi_{\{\beta_s\}} | e^{-i \vec{k} \cdot \vec{X}_L / \hbar} | \psi_{\{m_s\}} \rangle \quad (A.5)$$

M_C is the matrix element for the formation of the compound nucleus and M_{rad} is the matrix element for its decay. Both of these terms depend exclusively on the internal structure of the nucleus.

$\hat{X}_L$ is the position of the center of mass of the compound nucleus at L that we have singled out for this reaction. Since all the effects due to excitation of different nuclei are incoherent with each other it is legitimate to consider each nucleus separately and without a loss of generality.

$\Psi_{\{\alpha_s\}}$, $\Psi_{\{m_s\}}$ and $\Psi_{\{\beta_s\}}$ are the harmonic wave functions of the crystal in the initial, intermediate and final states, respectively. They are the harmonic oscillator wave functions since the lattice forces are assumed harmonic.

The matrix element $\langle \Psi_{\{m_s\}} | e^{+i\vec{p} \cdot \hat{X}_L / \hbar} | \Psi_{\{\alpha_s\}} \rangle$ is for the transfer of the momentum $\vec{p}$ of the incident neutron to the crystal and with the excitation of the lattice from an initial state to the intermediate state.

The matrix element $\langle \Psi_{\{\beta_s\}} | e^{-i\vec{k} \cdot \hat{X}_L / \hbar} | \Psi_{\{m_s\}} \rangle$ is for the transfer of momentum $\hbar\vec{k}$ to the lattice from the decaying nucleus with the lattice going from the intermediate state to the final state.

Since all lattice motions are to be expressed in terms of plane waves the interaction operators used in both of these matrix elements are necessary and unique since it is required that momentum be conserved. Any other interaction Hamiltonian will not do this.

Introducing these modifications then Eq. (A.1) takes the form

$$W(\{\beta_s\}; \{\alpha_s\}) = M_{\text{rad}}^2 M_c^2 \sum_{\{m_s\}} \frac{|\langle \Psi_{\{\beta_s\}} | e^{-i\vec{k} \cdot \hat{X}_L / \hbar} | \Psi_{\{m_s\}} \rangle \langle \Psi_{\{m_s\}} | e^{+i\vec{p} \cdot \hat{X}_L / \hbar} | \Psi_{\{\alpha_s\}} \rangle|^2}{[E - E_0 - \sum_s (m_s - \alpha_s) \hbar \omega_s]^2 + \frac{1}{4} \Gamma^2}.$$

(A.6)

In order to determine the total probability for neutron capture in addition to summing over all the intermediate states it is also necessary to sum over all the initial and final lattice states. Since the momentum transferred to the lattice during decay ($\hbar \vec{k}$) is much larger than the momenta components in the lattice, we can neglect the dependence of final states upon this momentum transfer. This approximation, however, is not valid for neutron capture since here the momentum transferred is much less. A plausible assumption made here is that the crystal is initially in thermal equilibrium so that the proper summing over these states is done by weighting them with the probability that the crystal is in the initial state $\{\alpha_s\}$. This probability is given by the notation $g(\{\alpha_s\})$ which is the Boltzmann weighting factor when the sample is at temperature T . Therefore, Eq. (A.6) takes the form

$$W(E) = \sum_{\{\beta_s\}} \sum_{\{\alpha_s\}} g(\{\alpha_s\}) \sum_{\{m_s\}} \frac{P(\{\beta_s\}; \{m_s\}) \left| \langle \psi_{\{m_s\}} | e^{i \vec{p} \cdot \vec{x}_L / \hbar} | \psi_{\{\alpha_s\}} \rangle \right|^2}{[E - E_0 - \sum_s (m_s - \alpha_s) \hbar \omega_s]^2 + \frac{1}{4} \Gamma^2} \quad (A.7)$$

where

$$P(\{\beta_s\}; \{m_s\}) = \left| \langle \psi_{\{\beta_s\}} | e^{-i \vec{k} \cdot \vec{x}_L / \hbar} | \psi_{\{m_s\}} \rangle \right|^2 \quad (A.8)$$

is the probability that the lattice will be in a particular final state $\{\beta_s\}$ after the transition from the intermediate state $\{m_s\}$. The matrix elements for M_{rad}^2 and M_c^2 have been dropped since they are common to all subsequent formulas and because interest now is on the lattice's effect on the resonance shape.

The first sum to be performed is over the final states since it only involves the P term. This sum, however, is unity, i.e.

$$\sum_{\{\beta_s\}} P(\{\beta_s\}; \{m_s\}) = 1, \quad (A.9)$$

since the intermediate state always returns to the final state.

Eq (A.7) therefore takes the form

$$W(E) = \sum_{\{\alpha_s\}} g(\{\alpha_s\}) \sum_{\{m_s\}} \frac{|\langle \Psi_{\{m_s\}} | e^{i \vec{p} \cdot \vec{X}_L / \hbar} | \Psi_{\{\alpha_s\}} \rangle|^2}{[E - E_0 - \sum_s (m_s - \alpha_s) \hbar \omega_s]^2 + \frac{1}{4} \Gamma^2} \quad (A.10)$$

The main problem in the development of this theory has to do with the interaction Hamiltonian $e^{i \vec{p} \cdot \vec{X}_L / \hbar}$ for the neutron with momentum $\vec{p}$ being incident on the nucleus at lattice position $\vec{X}_L$. Introducing normal coordinates it is possible to express the position vector $\vec{X}_L$ as

$$\vec{X}_L = \vec{X}_L^0 + \vec{U}_L \quad (A.11)$$

where $\vec{X}_L^0$ is the equilibrium position of the L^{th} atom and $\vec{U}_L$ is its displacement from equilibrium. Further, without loss of generality, the rest position of the atom is taken to be the origin of our coordinate system, i.e. $\vec{X}_L^0 = 0$ so that

$$e^{i \vec{p} \cdot \vec{X}_L / \hbar} = e^{i \vec{p} \cdot \vec{U}_L / \hbar} \quad (A.12)$$

The usual theory of lattice vibrations treated classically is based on the harmonic Hamiltonian where the potential energy function, expanded in powers of $\vec{U}_L$, can be terminated with the quadratic term. In this approximation the equations of motion for the nuclei can be solved to give the $3N$ normal modes of vibration, ω_s . In normal coordinates the crystal, treated as a system of simple harmonic oscillators, has a Hamiltonian which can be separated and

written as the sum of $3N$ independent harmonic Hamiltonians, i.e.

$$H = \sum_{s=1}^{3N} H_s \quad (\text{A.13})$$

where

$$H_s = \frac{1}{2M} P_s^2 + \frac{1}{2} M \omega_s^2 Q_s^2 \quad (\text{A.14})$$

P_s and Q_s are the normal momenta and coordinates, respectively. $M\omega_s^2$ is the force constant.

In this approximation Lamb is able to evaluate the matrix elements in Eq. (A.10) yielding:

$$\langle \psi_{\{\alpha_s\}} | e^{i \vec{p} \cdot \vec{x}_s / \hbar} | \psi_{\{\alpha_s\}} \rangle = \prod_s K(\alpha_s, \alpha_s; q_s) \quad (\text{A.15a})$$

where the propagation vector q_s is given by

$$q_s^2 = \frac{(\vec{p} \cdot \vec{e}_s)^2}{2M\hbar\omega_s N} \quad (\text{A.15b})$$

and where $\vec{e}_s$ is the unit polarization vector of the s^{th} normal mode. N is the large number of atoms in the crystal. Further it is shown that there exists only three terms in K with q_s^2 . They are

$$|K(\alpha_s, \alpha_s; q_s)|^2 = (1 - 2\alpha_s q_s^2) e^{-q_s^2}, \quad (\text{A.15c})$$

$$|K(\alpha_s + 1, \alpha_s; q_s)|^2 = (\alpha_s + 1) q_s^2 e^{-q_s^2}, \quad (\text{A.15d})$$

$$\text{and } |K(\alpha_s - 1, \alpha_s; q_s)|^2 = \alpha_s q_s^2 e^{-q_s^2}. \quad (\text{A.15e})$$

The K 's which may involve the creation or destruction of more than one phonon in a particular mode s are neglected in this approximation. It is assumed that because q_s^2 is inversely proportional to the number of atoms in the crystal, which is very large, higher order terms in q_s^2 may be neglected. If next the crystal's isotropy condition is applied, then averaging $\vec{e}_s$ for frequency ω_s over all

directions results in Eq. (A.15b) becoming

$$g_s^2 = \frac{R}{3\hbar\omega_s N} \quad (\text{A.15f})$$

where

$$R = \frac{p^2}{2M} \quad (\text{A.15g})$$

is the recoil energy of a free nucleus. In this limit then Eq. (A.10) becomes

$$W(E) = \sum_{\{\alpha_s\}} g(\{\alpha_s\}) \sum_{\{m_s\}} \frac{\prod_s |K(\dots)|^2}{[E - E_0 - \sum_s (m_s - \alpha_s)\hbar\omega_s]^2 + \frac{1}{4}\Gamma^2} \quad (\text{A.16})$$

In order to satisfy the energy conservation requirement a delta function dependence is introduced and the resulting expression is integrated over the entire range of energy transfer. $W(E)$ now takes the form

$$W(E) = \sum_{\{\alpha_s\}} g(\{\alpha_s\}) \sum_{\{m_s\}} \int_{-\infty}^{+\infty} \frac{\prod_s [\dots] \delta(\rho - \sum_s (m_s - \alpha_s)\hbar\omega_s) d\rho}{[E - E_0 - \rho]^2 + \frac{1}{4}\Gamma^2} \quad (\text{A.17})$$

where ρ is the energy transferred to the lattice. The next two steps in the development are (1) to write the delta function in integral form, and (2) to sum over all intermediate states where only three terms are possible differing from the initial state by 0, +1 or -1 phonon exchange. When these steps are completed $W(E)$ takes the form

$$W(E) = \frac{1}{2\pi} \sum_{\{\alpha_s\}} g(\{\alpha_s\}) \int_{-\infty}^{+\infty} d\rho \int_{-\infty}^{+\infty} \frac{du e^{iup}}{[E - E_0 - \rho]^2 + \frac{1}{4}\Gamma^2} \prod_s \left\{ 1 + g_s^2 [-2\alpha_s + (\alpha_s + 1) e^{-iu\hbar\omega_s} + \alpha_s e^{iu\hbar\omega_s}] \right\} \quad (\text{A.18})$$

where u is the dummy variable of the delta function integral representation.

In summing over the initial states the only information about $g(\{\alpha_s\})$ which is needed is the average value of the occupation number $\bar{\alpha}_s$ of each phonon state s . Since phonons obey Bose-Einstein statistics then for thermal equilibrium we use the distribution given by

$$\bar{\alpha}_s = \frac{1}{e^{\hbar\omega_s/kT} - 1} \quad (A.19)$$

Using these mean values results in the removal of the summation over the initial states and Eq. (A.18) becomes

$$W(E) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} d\rho \frac{e^{i u \rho}}{[E - E_0 - \rho]^2 + \frac{1}{4}\Gamma^2} \prod_s \left\{ 1 + g_s^2 \left[-2\bar{\alpha}_s + (\bar{\alpha}_s + 1) e^{-i u \hbar \omega_s} + \bar{\alpha}_s e^{i u \hbar \omega_s} \right] \right\} \quad (A.20)$$

The next step is to expand the product term. All contributions including terms higher than g_s^2 are dropped because of their smallness and this term is expressed in exponential form. $W(E)$ now takes the form

$$W(E) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} du \int_{-\infty}^{+\infty} d\rho \frac{e^{i u \rho} e^{g(u)}}{[(E - E_0) - \rho + \frac{i}{2}\Gamma]^2} \quad (A.21a)$$

where

$$g(u) = \sum_s g_s^2 \left[(\bar{\alpha}_s + 1) e^{-i u \hbar \omega_s} + \bar{\alpha}_s e^{i u \hbar \omega_s} - 1 - 2\bar{\alpha}_s \right] \quad (A.21b)$$

Performing the integration over ρ first by the method of residue gives

$$W(E) = \frac{2}{\Gamma} \mathcal{R} \int_0^{\infty} du e^{i u (E - E_0 + \frac{i}{2}\Gamma)} e^{g(u)} \quad (A.22)$$

(A.22)

where the real part is taken since the probability $W(E)$ is always a number.

Beyond Eq. (A.22) it is not possible to evaluate $W(E)$ exactly because of the complication caused by $g(u)$ in the exponential. Lamb, however, does reduce $W(E)$ further for two limiting cases: weak and strong binding.

The weak binding approximation is that $u\hbar\omega_s \ll 1$ which permits the exponential terms in $g(u)$ to be expanded and simplified to

$$g(u) = -iu \sum_s q_s^2 \hbar\omega_s - u^2 \sum_s q_s^2 \left(\bar{\alpha}_s + \frac{1}{2}\right) \hbar^2 \omega_s^2. \quad (\text{A.23})$$

In place of performing the summation over s , this operation is replaced by multiplying by the $3N$ independent modes of vibration. Applying Eq(A.15f) and recognizing that the average energy per degree of freedom is

$$\bar{E} = \left(\bar{\alpha}_s + \frac{1}{2}\right) \hbar\omega_s \quad (\text{A.24})$$

$g(u)$ takes the form

$$g(u) = -iuR - u^2 R \bar{E}. \quad (\text{A.25})$$

Also $W(E)$ becomes

$$W(E) = \frac{2}{\Gamma} \operatorname{Re} \int_0^\infty du e^{iu(E-E_0-R) - u\left(\frac{\Gamma}{2} + uR\bar{E}\right)} \quad (\text{A.26})$$

where the real and imaginary parts in the exponential have already been separated. Taking the real part of the integrand gives the result that

$$W(E) = \frac{2}{\Gamma} \int_0^\infty du \cos u(E-E_0-R) e^{-u\left(\frac{\Gamma}{2} + uR\bar{E}\right)} \quad (\text{A.27})$$

which indicates that the line form is not centered about the resonance energy but is shifted to a higher energy, $E = E_0 + R$.

Making the following substitutions

$$x = \frac{2(E - E_0 - R)}{\Gamma} \quad , \quad (A.28a)$$

$$y = \frac{u\Gamma}{2} \quad , \quad (A.28b)$$

$$\lambda = \frac{4R\bar{E}}{\Gamma^2} \quad , \quad \text{and} \quad (A.28c)$$

$W(E)$ becomes

$$W(E) = \frac{4}{\Gamma^2} \int_0^\infty dy \cos x y \exp(-y - y^2 \lambda) \quad (A.29)$$

Lamb indicates that the integral in Eq. (A.29) is equivalent to our Doppler function, $\Psi(x, \lambda)$, that we obtained in the ideal gas approximation. There is a difference, however, between a gas target and the weak binding limit for a solid and this difference is seen in Eq (A.28c) in the new definition of the Doppler width. Since for the gas the Doppler width is defined as

$$\Delta_{\text{gas}} = 2\sqrt{\frac{m}{M} k T E} \quad , \quad (A.30a)$$

whereas the crystal width,

$$\Delta_{\text{crystal}} = 2\sqrt{R\bar{E}} = 2\sqrt{\frac{m}{M} E \bar{E}} \quad , \quad (A.30b)$$

involves $\bar{E}$, the average energy per degree of vibration, in place of the sample's temperature, T . Assuming the Debye model is adequate to describe the crystal, Lamb relates the average energy to an effective temperature which replaces the sample's real temperature. In this limit one treats a solid as an ideal gas but at some higher temperature.

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