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FINAL REPORT
0412J0004-3G
TASK 26

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FINAL REPORT FOR TASK ORDER No. 26

Walter. J. Trela

Sumner Associates, Santa Fe

October, 1995

This is a final report for task order No. 26 between the University of California, Los Alamos National Laboratory and Sumner Associates issued 20 March, 1995. The task statement for order No. 26 is as follows:

"Provide assistance on the design of experimental arrangements for acquisition of data describing the interaction of neutrons with selected materials arranged in specific geometry."

"Provide assistance in the delivery of drawings and specifications associated with the task."

Three experimental descriptions follow; Dynamic Temperature Measurements in Explosives and Inert Simulants Using Neutron Resonance Radiography, Measurements of the Phonon Frequency Spectrum of Pu from Neutron Resonance Broadening, and Resonant Neutron Absorption Spectroscopy Of Hi-Temperature Superconductors, that fulfill the task statement.

WNR Research Proposal

(Use for nuclear physics experiments at either WNR or LANSCE. Return UNCLASSIFIED proposals to NPAC, MS H803, CLASSIFIED proposals to MS D406)

For NPAC use: Date received ___/___/___

Experiment Number _____

Experiment Title:

MEASUREMENTS OF THE PHONON FREQUENCY SPECTRUM OF Pu
FROM NEUTRON RESONANCE BROADENING

Proposal Status: ☐ New Proposal ☒ Continuation of Experiment No. 1N0042

NPAC Subcommittee:

☒ Classified and Weapons Physics

☐ Nuclear Technology

☐ Basic Nuclear Physics

Principal Investigator(s)/Affiliation/Address/Telephone/E-mail address: (underline local contact person)

<u>Walter Trela</u>	P-17/MS H803	5-4123
Angus Lawson	LANSCE/MS H805	7-8844
Eric Lynn	P-17/MS H803	7-1053

Collaborators/Affiliations/Address/Telephone/E-mail address:

Executive Summary of Proposal: (Please address motivation, objectives, scientific impact, and relevance to the Laboratory's mission, to allow proper evaluation of this submission. A guide to the key points is included. Add additional attachments as appropriate):

The experiment is to study by neutron transmission time-of-flight measurements at LANSCE the Doppler broadening of low energy neutron resonances of plutonium isotopes and a control material, probably tantalum, with the aim of obtaining information, in the form of frequency moments, of the phonon spectrum of plutonium metal in its delta-phase. Such information is fundamental to the basic understanding of the physical metallurgy of plutonium. This project has relevance to the Laboratory's weapons research; it should also add to the Laboratory's expertise and knowledge in materials technology in a more general way and have potential in study and design of new materials for industrial transfer.

The importance of these measurements with respect to science based stockpile stewardship was emphasized at the recent "Workshop on Defense, Basic, and Industrial Research at the Los Alamos Neutron Science Center".

Experimental Area:

- ☒ 1 (LANSCE) Flight Path 5
☐ 2 (Blue Room) Flight Path _____
☐ 4 (Target 4/WNR) Flight Path _____

Proton Beam Parameters:

Target 2: proton energy _____ beam spot size _____
macropulse spacing _____ micropulse spacing _____
protons/micropulse _____
Target 4: micropulse spacing _____

Resources needed: (data acquisition computer requirements, detector systems, shielding, samples/targets, technical support, signal cables, etc.)

Standard P-17 VAX Data acquisition system
1 Technician 1/4 time
Power supplies and other electronic equipment
The above are in addition to the standard equipment associated with FP 5

Special requirements: (nuclear materials handling, security guards, space required, cooling water etc.):

Displex (provided by LANSCE)

Time required: (assume LANSCE operation at 80 mA and WNR operation at specified beam conditions, include set-up and data collection time, please state in 8-hour shifts)

40 shifts

Scheduling restrictions: (availability of collaborators, timeliness of measurement, etc.):

August and September

Safety Issues:

☐ Toxic or corrosive material?	☒ Radioactive samples?
☐ Explosives or inflammable material?	☐ Induced radioactivity?
☐ Multiple entries requiring Health Physics	☐ Other? (Please explain)

DETAILED STATEMENT OF THE EXPERIMENT (Continued)

Knowledge of the phonon frequency spectrum $g(\nu)$ is required for basic theoretical understanding and calculations of the physical metallurgy of a material. Full data on the phonon spectrum can be obtained from neutron inelastic scattering measurements on a single crystal using, for example, a triple-axis spectrometer on a high flux research reactor. If suitable single crystals are not available, as in the case of plutonium, less complete information found in other ways will be very valuable. Our proposal is to study the Doppler broadening of narrow low energy neutron resonances of plutonium isotopes over a range of temperatures and in sufficient detail to extract some of the low-order moments and "quasi-moments" of the phonon frequency spectrum. ("Quasi-moments" are weighted by temperature dependent functions of the frequency). In these experiments the samples can be metallic foils or cylinders in normal micro-crystalline form, or even powders. It should be noted that Doppler broadening gives phonon information specific to the frequencies residing on the atom with the resonance cross section. Thus the method, if successful, could be applied to a range of alloys and other materials with suitable elemental constituents containing narrow neutron resonances, and hence contribute to the Laboratory's expertise in advanced materials technology for advanced industrial projects as well as weapons research. This project should also greatly improve methods of treating Doppler broadening in nuclear resonance analysis, and thus contribute to the accuracy of the nuclear data base.

In the crudest approximation Doppler broadening is treated in a classical gas model [1]. This gives rise to an effective cross-section in which the true nuclear cross section in the centre-of-mass frame is convoluted with a Gaussian function (of the energy transferred to or from the surrounding medium) with dispersion parameter related to the average recoil energy $\bar{E}$ given to the compound nucleus from the momentum of the bombarding neutron and the ambient temperature of the target material. In the next approximation [2], for a crystalline target, the temperature in the dispersion parameter is replaced by the mean energy of occupied oscillator states; this is the "quasi-moment" (of order 1)

$$\langle h\nu \rangle_T = \int d\nu h\nu g(\nu) \coth(h\nu/2kT) \quad (1)$$

In practice, this approximation works quite well for neutron resonances measured at normal room temperatures and above, the difference between $\langle h\nu \rangle_T$ and kT being a few percent or less. Higher order approximations can be achieved by adding to the Gaussian convolution function extra terms that are the products of Hermite polynomials (in the energy transfer to the crystal lattice) with the Gaussian. The coefficients of these can be expressed in terms of the moments and "quasi-moments" of the phonon frequency spectrum. Beyond the unadorned Gaussian, one may expect the "skew" and "kurtosis" terms (involving third and fourth order polynomials, respectively) to be significant. The coefficients of these terms can be expressed using the second phonon moment $\langle (h\nu)^2 \rangle$ and the quasi-moment of order 3, $\langle (h\nu)^3 \rangle_T$.

Such expansions of the convolution function can be very accurate with a reasonably small number of terms (fewer than ten) except for resonances below a few eV. At these low energies a substantial fraction of the resonance cross section resides in a recoilless "line", the neutron analogue of gamma-ray Mossbauer spectra. The fraction of the resonance cross section in this recoilless profile is given by the Debye-Waller factor

$$P = \exp[-\bar{E} \langle (h\nu)^{-1} \rangle_T] \quad (2)$$

The recoilless line arises from a delta function in the Doppler-broadening convolution function. The delta function is not amenable to a treatment based on moments. Hence such cases are best treated by first separating out the recoilless line and then treating the remainder of the convolution function by the moments method. It should be noted, however, that the Debye-Waller factor of eq. 2 contains the quasi-moment of order -1, and hence also gives important information on the phonon spectrum. Indeed, this quantity has been extracted for this purpose from powder diffraction measurements in thermal neutron scattering [3].

It is the objective of this proposal to determine the first 3 or 4 moments (or quasi-moments) of the phonon spectrum of plutonium metal, stabilised in its delta phase by alloying with a few percent of lighter elements, from accurate measurements of the low energy neutron resonance total cross sections of the plutonium isotopes at different temperatures. The cross sections are to be measured by the transmission method. Figure 1 shows simulated cross sections of a typical resonance (the 2.68 eV resonance of ^{242}Pu) for three fictitious phonon spectra having the same Debye-Waller factor but different higher order moments. This demonstrates that cross section differences of only a few percent can carry important information, and hence our aim is to measure transmission with a statistical accuracy better than a few tenths of a percent. Total detected neutron count accumulations per energy point interval are therefore required to be of the order of 10^6 . Neutron resolution must be high, preferably better than about one tenth of the resonance natural width. Measurements at the 60 m flight path station on flight path 5 at LANSCE, should achieve this quality of resolution for resonances of 10 eV or less. On this flight path the required count accumulation for one sample of 1 cm² cross sectional area can be achieved in about 8 hours.

To make accurate measurements over resonance peaks and well down on to their wings, and to cover resonances of widely different peak cross-section, a broad range of sample thicknesses will have to be used.

For some resonances (like that illustrated in Fig.1) the sample would be impractically thin if used in the pure isotopic form. Hence we shall exploit thicker samples of nominal Pu-239 and Pu-242 with impurities of the other isotopes at the few % level to obtain these data. We shall require runs on at least 5 samples ranging from about 0.1 mm to several cm to acquire useful data on the several candidate resonances for these studies. Temperature values will range from liquid helium to room. For all these runs some 200 h of beam time will be required. In addition it would be desirable to make similar measurements for control purposes on a metal such as tantalum for which the phonon characteristics are well known. However, these measurements could be made with samples of much larger cross sectional area, so the beam time requirements would be much less.

References

- [1] H A Bethe, Rev.Mod.Phys. 9,140 (1937)
- [2] W E Lamb, Phys.Rev.55,190 (1939)
- [3] G H Kwei, A C Lawson, M Mostoller, Physica C175,135 (1991)

Figure Caption

Calculated effective cross sections over the 2.68 eV neutron resonance of ^{242}Pu at temperature 4K. The three phonon spectrum models are fictitious, for illustrative purposes only. They have the features that the Debye-Waller factors are identical, with $\langle 1/\hbar\nu \rangle = 66.5 \text{ eV}^{-1}$. For the squares, $\langle \hbar\nu \rangle = 16.7 \text{ meV}$, $\langle (\hbar\nu)^2 \rangle = 301 \text{ meV}^2$, for the crosses, $\langle \hbar\nu \rangle = 17.9 \text{ meV}$, $\langle (\hbar\nu)^2 \rangle = 370 \text{ meV}^2$, for the stars, $\langle \hbar\nu \rangle = 25.2 \text{ meV}$, $\langle (\hbar\nu)^2 \rangle = 734 \text{ meV}^2$.

Pu-242+n; 2.68 eV resonance; T=4K

WNR Research Proposal

(Use for nuclear physics experiments at either WNR or LANSCE. Return UNCLASSIFIED proposals to NPAC, MS H803, CLASSIFIED proposals to MS D406)

For NPAC use: Date received / /

Experiment Number

Experiment Title: Dynamic Temperature Measurements in Explosives and Inert Simulants Using Neutron Resonance Radiography

Proposal Status: ☒ New Proposal ☐ Continuation of Experiment No.

NPAC Subcommittee:

☒ Classified and Weapons Physics ☐ Nuclear Technology ☐ Basic Nuclear Physics

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Executive Summary of Proposal: (Please address motivation, objectives, scientific impact, and relevance to the Laboratory's mission to allow proper evaluation of this submission. A guide to the key points is included. Add additional attachments as appropriate):

Motivation: Localized temperature excursions ("hotspots") in explosives subjected to shock, shear, or friction control ignition and influence the evolution of reaction and are thus critical to safety, reliability, and performance of explosives in nuclear and conventional weapons. Such temperature excursions are estimated to be a few to many hundreds of degrees Celsius, depending upon conditions and depending upon the modeling assumptions. Accurate modeling of transient explosive behaviors requires accurate prediction of hotspot temperatures. There are no reliable direct measurements of these temperature excursions.

Objectives: Demonstrate the capability and obtain direct measurements of temperatures generated in dynamic experiments on explosives.

Scientific Impact: Dynamic experiments using a pulsed neutron source have never been performed before. Conventional methods of measuring temperatures (thermocouples, optical pyrometry, infrared imaging) in explosive experiments have been only partially successful. Thermocouples perturb the flow, usually have inadequate rise times for shock experiments, and have questionable calibrations in the time/temperature domains of interest. Both pyrometry and infrared imaging techniques are useful for surface measurements only, and have problems with the validity of the underlying assumptions. The neutron resonance radiographic technique will provide the first accurate, internal, direct measurements of temperatures in dynamic explosives experiments.

Relevance: The experiments planned will provide the first accurate measurements of temperatures under conditions leading to initiation of explosive reactions. Accurate measurements of temperatures under these conditions will provide data critical to the construction and validation of models of initiation currently under development and badly needed by the weapons community for assessment of safety, reliability, performance of conventional and nuclear weapons.

Experimental Area:

- ☐ 1 (LANSCE) Flight Path 5
- ☐ 2 (Blue Room) Flight Path _____
- ☐ 4 (Target 4/WNR) Flight Path _____

Proton Beam Parameters:

Target 2: proton energy _____ beam spot size _____
macropulse spacing _____ micropulse spacing _____
protons/micropulse _____

Target 4: micropulse spacing _____

Resources needed: (data acquisition computer requirements, detector systems, shielding, samples/targets, technical support, signal cables, etc.)

Flight path 5, lithium 6 glass detector, flight path 5 data acquisition system.

Special requirements: (nuclear materials handling, security guards, space required, cooling water etc.):

We will be bringing small amounts of explosive into the area. We must be able to work in the area while the beam is on to set up experiments.

Time required: (for 800 MeV proton beam, assume WNR operation at 100 Hz, 3×10^8 p/micropulse, with micropulse spacing specified above; Target 2 operation at other energies will be at 12 Hz. Given in units of 8-hour shifts)

63 units, broken into at least two blocks.

Scheduling restrictions: (availability of collaborators, timeliness of measurement, etc.):

None. We would prefer to have one block of time shortly after beam time first becomes available (late-August), a second block of time towards late September.

Safety issues: ☐ Toxic or corrosive material? ☐ Radioactive samples?
☒ Explosives or inflammable material? ☐ Induced radioactivity?
☐ Multiple entries requiring Health Physics ☐ Other? (Please explain)

WNR/LANSCE NPAC Research Proposal for LAMPF Cycles 69-70 Page 2 of 2

DETAILED STATEMENT OF THE EXPERIMENT

Experiment Title: Dynamic Temperature Measurements in Explosives and Inert Simulants Using Neutron Resonance Radiography

Address the following in detail: Motivation, objectives, scientific impact, and relevance to the Laboratory's mission, relevance to other experiments and theory, pertinent references, description of experiment and techniques to be used, measurements to be made, data rates and possible backgrounds, results and precision looked for, evidence of ability of research team to carry out the experiment. If the proposal is for a continuation of work already approved by the NPAC, attach a progress report using the Experiment Report Form. Use front and back of this sheet if necessary.

Motivation: Localized temperature excursions ("hotspots") in explosives subjected to shock, shear, or friction control ignition and influence the evolution of reaction and are thus critical to safety, reliability, and performance of explosives in nuclear and conventional weapons. Such temperature excursions are estimated to be a few to many hundreds of degrees Celsius, depending upon conditions and depending upon the modeling assumptions. Accurate modeling of transient explosive behaviors requires accurate prediction of hotspot temperatures. There are no reliable direct measurements of these temperature excursions. We are currently taking several different approaches to calculating these temperatures. Current modeling includes the effects of frictional heating, thermal softening, melting, heat of fusion, heat transfer, and viscous heating. Unfortunately, the calculated temperatures are quite sensitive to the input parameters, and there is very little relevant data regarding these input parameters.

A similar situation exists for explosives equation of state predictions. Theoretical efforts to construct realistic and accurate equations of state extend back over fifty years. Variables amenable to direct measurement in dynamic explosive experiments are pressure, particle velocity, and shock velocity. From these measurements and initial state measurements, one can construct an incomplete equation of state. A missing critical piece of information is knowledge of the temperature. Direct (and accurate) measurement of the temperature would allow construction of a realistic complete equation of state.

Objectives: Demonstrate the capability and obtain direct measurements of temperatures generated in dynamic experiments on explosives.

Scientific Impact: Dynamic experiments using a pulsed neutron source have never been performed before. Conventional methods of measuring temperatures (thermocouples, optical pyrometry, infrared imaging) in explosive experiments have been only partially successful. Thermocouples perturb the flow, usually have inadequate rise times for shock experiments, and have questionable calibrations in the time/temperature domains of interest. Both pyrometry and infrared imaging techniques are useful for surface measurements only, and have problems with the validity of the underlying assumptions. The neutron resonance radiographic technique will provide the first accurate, internal, direct measurements of temperatures in dynamic explosives experiments.

Relevance: The experiments planned will provide the first accurate measurements of temperatures under conditions leading to initiation of explosive reactions. Accurate measurements of temperatures under these conditions will provide data critical to the construction and validation of models of initiation currently under development and badly needed by the weapons community for assessment of safety, reliability, performance of conventional and nuclear weapons.

Approach: We plan to use neutron resonance radiography to make accurate temperature measurements in explosives under a variety of important experimental conditions. The experiments performed this year will provide absolutely critical input to ongoing modeling efforts and will lay the groundwork for additional experiments to be performed in outlying years. In each experiment, we plan to use a heavy metal dopant to provide a strong resonance peak. The material of choice at present is gold, because of its extremely high resonance peaks at low neutron energies. Temperatures will be inferred from Doppler line broadenings.

A. Calibration and proof of principle experiments: In order to demonstrate the accuracy of the neutron radiography technique for this particular application, we plan to coat one cm and one and one half cm square substrates with a gold layer approximately 5 microns thick. (Calculations indicate that these are the minimum areas and gold thicknesses required to generate reasonable strength signals. For ease of handling and for safety considerations, we want to use the smallest reasonable amounts of explosives in each experiment.) Samples will be placed within an oven, with thermocouples, and heated to temperatures in the 200-1000 degree Celsius range. Neutron resonances signals at four different temperatures will be obtained. We will measure the neutron resonances in single-shot and multi-shot mode as a function of temperature to understand fully the limitations of single-shot experiments. Data will be used to infer the accuracy of the use of Doppler shifts in neutron resonances for temperature measurements in the multi microsecond time frame.

B. Shear and Friction Experiments: We plan to conduct a series of experiments to measure the shear and frictional heating of explosives. The friction experiments will involve the rapid sliding of an explosive sample across an inert substrate, at a specified sliding velocity and applied load. In the case of shear experiments, the setup and procedures are similar, except that the experiment is modified to force shear on a surface internal to the explosive, rather than at a substrate - explosive interface. Sufficient experiments are planned to allow mapping of temperature- time profiles as a function of sliding velocity and applied load. The intention is to measure temperatures close to the ignition threshold, although ignition itself is not a requirement of the experiment. The experiments will use small (of the order of a gram or two) samples of explosives. These small samples, properly configured, will be large enough to allow a reasonably strong signal generation from the gold, yet will be sufficiently small that they present no hazard to the laboratory setting and instrumentation, in the event of detonation. (We are in the process of initiating the formal safety review required for all new explosives experiments.) For shear and frictional heating experiments we are considering two approaches to introduction of the gold: In one case, we will vapor deposit the gold onto an explosive (PBX 9501) surface, and then mate that surface to another piece of explosive. In the second case, we will vapor deposit gold onto particles of HMX, from which we will form a molding powder which will be used with standard HMX molding powder to form a compact containing a striation of gold - coated material. The explosive samples are then placed into a holder of high strength aluminum, and coupled to a specially designed gun to produce reproducible sliding velocities. Use of these two approaches will help us in assessing the effect the gold has in perturbing the strain localization and thermalization processes in the experiment. Also, each experiment we plan to do at LANSCE will be performed many times in our own laboratory setting prior to implementation at LANSCE. This will allow us to work out additional timing, safety, and ancillary instrumentation issues well in advance.) We are currently designing and will use a fixture which will allow us to vary the sliding velocity and the pressure independently, and to time the experiments to the LANSCE beam with a high degree of precision. In our laboratory, we are setting up this experiment to obtain temperature measurements at an explosive - window interface using four color pyrometry and light pipes and an infrared imaging camera. We are also using a laser speckle interferometer to measure dynamic strain field evolution at the same time. (The major difficulty with these experiments performed in our laboratory is that they are contaminated with interface and three dimensional effects. The neutron resonance radiography approach allows us to sample the interior and to eliminate the interface problem and minimize the dimensionality problem.)

WNR Research Proposal

(Use for nuclear physics experiments at either WNR or LANSCE. Return UNCLASSIFIED proposals to NPAC, MS H803, CLASSIFIED proposals to MS D406)

For NPAC use: Date received / /

Experiment Number

Experiment Title:

RESENANT NEUTRON ABSORPTION SPECTROSCOPY OF HI-TEMPERATURE SUPERCONDUCTORS

Proposal Status: ☒ New Proposal ☐ Continuation of Experiment No. 1N0041

Proposal Type: ☐ Development ☐ Shared Operation ☒ Sole Use

Proposal Content: ☐ Classified and Weapons Physics ☐ Nuclear Technology ☐ Basic Nuclear Physics

Principal Investigator(s)/Affiliation/Address/Telephone: (underline local contact person)

Trela, Walter P-17 MS H803 5-4123

Lynn, Eric P-17 MS H803 7-1053

Collaborators/Affiliations/Address/Telephone:

Kwei, George LLNL MS H805 7-8840

Executive Summary of Proposal: (Please address motivation, objectives, scientific impact, and relevance to the Laboratory's mission, to allow proper evaluation of this submission. A guide to the key points is included on the second page. Add additional attachments as appropriate):

The study of the high- T_c oxide superconductors has yet to reveal the mechanism responsible for their superconductivity. However, a large accumulation of experimental and theoretical work on the high T_c superconductors suggests the importance of the electron-phonon interaction, as is the case for BCS superconductors. But, these results do not prove that phonons are directly connected to the mechanism for superconductivity.

We propose to continue the study of the lattice dynamics of high temperature superconducting materials by performing Doppler broadening neutron resonance spectroscopy on high T_c materials that have structural properties and phonon characteristics that suggest a strong coupling to the superconducting transition. Experiments will continue to concentrate on LaBaCuO₄ materials.

- ☒ 1 (LANSCE) Flight Path 5
☐ 2 (Blue Room) Flight Path _____
☐ 4 (Target 4/WNR) Flight Path _____

Proton Beam Parameters:

Target 2: proton energy _____ beam spot size _____
macropulse spacing _____ micropulse spacing _____
protons/micropulse _____
Target 4: micropulse spacing _____

Resources needed: (data acquisition computer requirements, detector systems, shielding, samples/targets, technical support, signal cables, etc.)

Standard P-17 VAX Data acquisition system
1 Technician 1/4 time
Power supplies and other electronic equipment

The above are in addition to the standard equipment associated with FP-5

Special requirements: (nuclear materials handling, security guards, space required, cooling water etc.):

Display refrigerator

Time required: (assume LANSCE operation at 80 mA and WNR operation at specified beam conditions, include set-up and data collection time, please state in 8-hour shifts)

40 shifts

Scheduling restrictions: (availability of collaborators, timeliness of measurement, etc.):

August and September

Safety Issues: ☐ Toxic or corrosive material? ☐ Radioactive samples?
☐ Explosives or inflammable material? ☒ Induced radioactivity?
☐ Multiple entries requiring Health Physics ☐ Other? (Please explain)

RESONANT NEUTRON ABSORPTION SPECTROSCOPY OF HI-TEMPERATURE SUPERCONDUCTORS

The study of the high- T_c oxide superconductors has yet to reveal the mechanism responsible for their superconductivity. However, a large accumulation of experimental and theoretical work on the high T_c superconductors suggests the importance of the electron-phonon interaction, as is the case for BCS superconductors. But, these results do not prove that phonons are directly connected to the mechanism for superconductivity.

We propose to continue the study of the lattice dynamics of high temperature superconducting materials by performing Doppler broadening neutron resonance spectroscopy on high T_c materials that have structural properties and phonon characteristics that suggest a strong coupling to the superconducting transition. Experiments will continue to concentrate on LaBaCuO₄ materials.

The lattice dynamics of the high T_c superconductors have been studied by a number of techniques. These include Raman scattering, ion channeling, x-ray absorption neutron inelastic scattering and neutron resonance absorption spectroscopy. But, the high-temperature superconductors are complex materials that contain a number of elements so that the vibrational spectra consist of overlapping contributions from the modes of all the constituents. Neutron resonance absorption spectroscopy (NRAS) has the great advantage that the vibrations can be studied separately for each element in the superconductor. Furthermore, in a non isotropic sample that can be oriented in the neutron beam the lattice dynamics can be studied in different directions in the sample. Also, NRAS can be very precise and $\langle E \rangle$ has been determined in some experiments to within 5 K, with even better accuracy at low temperature. These advantages have permitted the observation of the softening in the phonon density of states at the superconducting transition for the first time in Bi₂Sr₂CaCu₂O₈ (1).

Our proposal is to continue to study the Doppler broadening of narrow low energy neutron resonances (NRAS) of certain of the constituent elements in other high T_c materials, namely the LaBaCuO compounds. These measurements will be over a range of

temperatures above and below the superconducting transition temperature and in sufficient detail to extract some of the low-order moments and "quasi-moments" of the phonon frequency spectrum. As already noted, the Doppler broadening gives phonon information specific to the frequencies residing on the atom with the resonance cross section (See the attached appendix for a detailed discussion of the NRAS technique).

SUMMARY REPORT ON TASK 26 OF B.O.A. BETWEEN LANL AND SUMNER ASSOCIATES

J Eric Lynn

Abstract

Task 26 relates to applications of neutron resonance Doppler broadening. For LANL important possible areas of application include dynamic temperature measurement and phonon characteristics of materials, especially plutonium. The theory of Doppler broadening has been developed and computer programs written to allow design and analysis of experiments in this field. A paper is in process of publication by Nuclear Instruments and Methods on theory and potential of the method. A survey has been made of the most suitable elements, isotopes and resonances that can be used as doping agents for dynamic temperature measurement. The likely measurement accuracy that can be achieved with these agents for single pulse measurements based on the LANSCE neutron source has been calculated. It has been shown that accuracies of better than 10% and in some cases better than 5% using foils of only tens of microns thickness can be achieved in principle. In the area of phonon measurement effort has been concentrated on study and analysis of existing data (taken in the 1993 LANSCE season). This study has revealed problematic features associated with the neutron detector and the moderator-reflector assembly at the neutron source. Both of these have long time attenuation components in their response functions, and hence adversely affect resolution. By analysis of many resonances for several different materials of varying thickness the main characteristics of the detector response have been determined. The moderator time response function has been analysed from Monte Carlo calculations supplied by LANSCE staff. With the reasonably good information on the resolution function thus obtained, analysis of the resonance data on tungsten, tantalum and plutonium metals has been carried out to obtain physics information. It has been shown from the tungsten and tantalum data that sound information on the moments of the phonon spectrum can be obtained. Although the plutonium sample with which we were supplied was far from ideal, qualitatively reasonable phonon information resulted. Improvements that need to be made in future work include much better samples of plutonium, better thermometry, including direct measurement of sample temperature, and new methods of analysis to cancel remaining deficiencies in our knowledge of the resolution function.

I. Introduction: theory and applications of Doppler broadening

The slow neutron cross sections of most heavy nuclides have resonances in their energy dependence that are narrow enough to be significantly Doppler broadened. The detailed nature of the Doppler broadening depends on the environment of the nucleus - the solid, liquid or gaseous state, and temperature, in which it is embedded. Information on the nuclear environment can therefore be extracted by detailed measurement of the form of the neutron cross section in the resonance region.

The basic papers are those by Bethe [1], on Doppler broadening in a gaseous medium, and Lamb [2] on the basic formalism for crystals. We have developed the work of the latter to produce expansions for the Doppler broadening function that depend on the moments of the phonon spectrum of the crystal (or, in a multi-atomic compound or alloy, the fraction of the spectrum that resides on the nucleus carrying the resonance). The effective cross section that can be measured for a nucleus in its material environment is

$$\sigma_{\text{eff},t} = \int dE' S(E') \sigma_t(E_n - E') \quad (1)$$

where σ_t is the true neutron cross section (for a free, stationary nucleus), and $S(E')$ is an energy-transfer function (the Doppler broadening function) governing the transfer of energy with the environment of the nucleus in the neutron absorption process. We have shown that a useful and accurate expression for this function is

$$S(E') = \frac{1}{\Delta\sqrt{\pi}} \exp[-(E' - R)^2/\Delta^2] \left[1 + \sum_{n=3}^{\infty} a_n H_n[(E' - R)/\Delta] \right] \quad (2)$$

where R is the recoil energy that would be carried off by the compound nucleus after absorption of the neutron by a free, stationary target nucleus, the H_n are the Hermite polynomials and the "Doppler width" for a crystalline sample is

$$\Delta = \sqrt{2R\langle h\nu \rangle_T}, \quad (3)$$

$$\langle h\nu \rangle_T = \int d\nu \, h\nu g(\nu) \coth(h\nu/2kT) \quad (4)$$

$g(\nu)$ is the spectrum of phonon frequencies carried by the crystal, k is Boltzmann's constant and T the sample temperature. The coefficients a_n are functions of the simple even moments of the phonon spectrum, the odd moments with the additional Boltzmann weighting factor (as in eq.4) and the recoil energy. The first two terms within the summation of eq.2 have the coefficients

$$a_3 = \langle (h\nu)^2 \rangle / 12\Delta \langle h\nu \rangle_T \quad (5)$$

$$a_4 = \langle (h\nu)^3 \rangle_T / 96R \langle h\nu \rangle_T^2 \quad (6)$$

The full theoretical development and equations for the coefficients are to be found in a paper in process of publication in Nuclear Instruments and Methods B, by J.E.Lynn and W.J.Trela [3].

The principal parameter that can be determined by measurements of the effective cross section is Δ , the Doppler width, and hence the first Boltzmann-weighted moment $\langle h\nu \rangle_T$. At very low temperatures this becomes simply the mean energy of the phonon spectrum. Also at low temperatures the additional terms with coefficients a_3 , a_4 , marking departures in skewness and kurtosis from the simple gaussian, are significant for low energy resonances and hence can give information on the second and third moments of the phonon spectrum. Such phonon information, specific to the resonant atomic type, can be valuable for theoretical metallurgy and material physics [4].

At high temperatures (generally speaking, room temperature and above) the "moment" $\langle h\nu \rangle_T$ approximates to the temperature,

$$\langle h\nu \rangle_T = 2kT + \langle (h\nu)^2 \rangle / 6kT + \dots \quad (7)$$

while the coefficients a_n of the terms beyond the simple gaussian of eq.2 become very small. Eq.2 is then approximated by the classical gas formula of Bethe [1]. In this regime Doppler broadening can be used as a technique for temperature measurement, with the possibility, for a sufficiently intense pulsed neutron source, of a "dynamic" measurement of temperature during a transient event with a time duration as short as a few micro-seconds. Many materials may not have elements with suitable resonances. In such cases trace elements can be introduced, and the spatial position of such introductions allows an element of radiography.

In this report we summarize our analytical investigations to date on the use of the LANSCE neutron source for Doppler broadening studies of both dynamic temperature measurement (section II) and phonon properties (section III).

II. Resonance neutron methods for dynamic temperature determination: some dopant elements and achievable accuracies

Most heavy nuclides (mass number greater than about 60) have resonances in their neutron cross sections that are narrow enough to suffer significant Doppler broadening. The Doppler width increases with neutron energy through the recoil energy (eq.2). For low energy resonances the radiation width, which does not have a significant energy dependence, is greater than the neutron width, which does. Hence, as a general rule, the Doppler width will be greater relative to the total resonance width for resonances at higher neutron energy. This does not imply, however, that the high energy resonances are better for temperature determination. Other factors lead to the opposite conclusion. Firstly, energy resolution deteriorates much faster than the Doppler width increases. Secondly, the neutron intensity (per unit energy interval) from a moderated pulsed neutron source is greater for low energies. More detail is presented in Appendix A.

Experiments on dynamic temperature measurement using observation of Doppler broadening of neutron resonances in a single pulse of neutrons from the LANSCE source are presently an important research area for stockpile stewardship. The high explosive materials under study do not have suitable nuclear properties for temperature determination, and therefore must be doped. An important issue is the choice of element as a doping material for the temperature determination. Elements with strong (to minimize the required amount of dopant), narrow

resonances in their neutron cross-sections are required. We have surveyed a number of elements that are potentially useful in this respect. For this survey, calculations were based on the normal neutron intensity of the LANSCE source, the efficiency of an available lithium glass neutron detector operating in current-mode and an assumed sample (and beam) area of 1cm^2 . Computer programs were written to create simulated data files of neutron transmission data through resonances, and to carry out least-squares analysis to determine temperature and associated error as a standard deviation. See Appendix A.

The elements surveyed include indium, samarium, dysprosium, hafnium, tantalum, gold, uranium-234,238, plutonium-240,242. Sensitivity depends on the thickness of the layer of element. All these elements were studied over the range of thickness 0.165×10^{-4} to 0.99×10^{-4} atoms per barn (10^{-24}cm^2). These correspond, in gold, for example, to between 3 and 17 μm of metal. At room temperature possible accuracies of better than 10% can be achieved. Indium, because of its low energy resonance at 1.45eV, will give 9% accuracy with a 25 μm foil ($0.99 \times 10^{-4} \text{ a.b}^{-1}$). Only the transuranics will do better. At higher temperatures higher accuracies can be achieved. Indium remains good in principle (4.2% accuracy at 1200K with the 25 μm foil) but its low melting point becomes a problem. Other elements approach this accuracy. Thulium (melting point greater than 1800K) gives 5.8% accuracy from its 3.91 eV resonance. Samarium and hafnium give slightly poorer accuracies (~9%) in their natural elemental composition, but as separated isotopes (^{152}Sm , resonance at 8.05 eV; ^{178}Hf , 7.78eV) they offer the possibility of achieving this with considerably thinner foils. If HSE issues could be overcome, plutonium-240 with its resonance at 1.06eV offers approximately twice the accuracy of any other nuclide. A fuller description of this study is given in Appendix A.

III. Phonon spectrum moments

A. Instrumental problems

The first experimental data on resonance Doppler broadening at low temperatures were taken at LANSCE in 1993 by W J Trela and K Meggers. The experimental facility consisted of Flight Path 5 with a neutron detector, supplied by H Priesmeyer of Kiel University, Germany, at 56 m. This facility was originally designed and used for the study of Bragg edges at very low (meV energies) in the neutron effective total cross section. The detector is a large area lithium-loaded scintillating glass operating in the current-mode [5] to accept very high count rates. The new use of this facility in the resonance (eV) region implied that a new set of instrumental problems would be encountered. These are associated mainly with resolution and background.

Irreducible factors in resolution broadening are the proton pulse length delivered from the PSR (this is 0.125 μs full-width at half-maximum), the pulse broadening of neutrons of a specific energy due to the moderator and reflector assembly around the tungsten proton target and the rise time of the response of the lithium glass scintillator detector (about 0.08 μs for its fastest component). The timing channel length can be adjusted to the requirement of the experiment.

The background is usually determined by the "black" resonance method; materials with strong resonances are placed in the beam in foils of sufficient

thickness that no neutrons penetrate for a significant energy range around the peak. It was soon noticed with this technique that the background at each resonance apparently had a rate of energy variation that was much greater than that between different resonances. This implied a component in the resolution function with a very long time delay (several μs).

1. Moderation time spread

One possible source of this is the moderation time spread. Our initial information on this was simply the variance of this time spread as calculated by G. Russell using Monte Carlo techniques for his design of the moderator-reflector assembly at the LANSCE target [6]. His value for the square-root variance is (in μs) $2.8 E(\text{in eV})^{-0.394}$. More detailed Monte Carlo calculations in the form of time-binned histograms for neutron energies between 1 and 20 eV were supplied by Luc Daemen of LANSCE. The expected form of the time-spread for a simple slab moderator is

$$p(t) = (t^2/2\tau)\exp(-t/\tau) \quad (8)$$

This is a member of the so-called χ^2 family of functions, with ν parameter ("number of degrees of freedom") equal to 6. It does not describe the very long tail apparent in L. Daemen's calculations. We have established that a function that results from convolution of a gaussian with an exponential is also inadequate for representing the Monte Carlo results. Hence we attempted to fit these data with a function that is the sum of two χ^2 functions, one, describing mostly the main peak, having $\nu=6$, and the other, for the tail, with variable ν , which turned out to be best fitted with $\nu=3.5$. The overall function that was fitted to the Monte Carlo data is therefore

$$p(t) = w_1 f_1(t) + (1-w_1) f_2(t) \quad (9)$$

where

$$\begin{aligned} f_1(t) &= [(t-t_1)^2/2\tau] \exp[-(t-t_1)/\tau_1], & t \geq t_1 \\ &= 0, & t < t_1 \\ f_2(t) &= [(t-t_2)^{0.75}/0.3756\Gamma(1.75)\tau_2] \exp[-1.75(t-t_2)/\tau_2], & t \geq t_2 \\ &= 0, & t < t_2 \end{aligned}$$

The best-fit parameters found for this function are:

$$\tau_1(\text{in } \mu\text{s}) = 0.7404/\sqrt{E(\text{in eV})}$$

$$t_1(\text{in } \mu\text{s}) = 0.4908/\sqrt{E(\text{in eV})}$$

$$w_1 = 0.755$$

$$\tau_2(\text{in } \mu\text{s}) = 4.027/\sqrt{E(\text{in eV})}$$

$$t_2(\text{in } \mu\text{s}) = 3.847/\sqrt{E(\text{in eV})}$$

The square root variance of this function is $4.54/\sqrt{E}$. The function of eq.9, with the above parameter values, very well describe Daemen's Monte Carlo data over a wide range of time.

2. Phosphorescent decay

The attenuation length of the tail component of the moderation spread function is insufficient to explain the slope of the apparent local background at black resonances, especially at higher energies where it is relatively unchanged. It has been suspected, however, that the lithium glass scintillator emits some of its light in at least one component decaying with long life ($\sim 10\mu\text{s}$) [5]. We have therefore analysed transmission data for a range of resonances with energies of about 10eV to above 100eV in tungsten and tantalum from foils with a range of thicknesses. We find, on this hypothesis, that approximately 36% of the light is emitted in a component with attenuation length of $9.2\mu\text{s}$. We have used these figures in our analyses of low energy resonances in tungsten, tantalum and plutonium to obtain phonon data.

B. Results of resonance analysis

We have established, in ref.[3], by numerical simulation and analysis, that it is possible with high intensity pulsed neutron sources to determine the first, Boltzmann-weighted moment with better than 0.5% accuracy, the second moment with about 2% accuracy, and the third (weighted) moment with better than 10% accuracy. With samples of suitable thickness, these moments can be determined independently of the resonance nuclear parameters, which also emerge from least square analysis with high accuracy. We have now analysed real experimental data taken with the LANSCE flight path 5 facility by Trela and Meggers in 1993. The main aim of the experiments was to obtain phonon spectrum information on plutonium metal. Experiments were also performed on tungsten and tantalum metals, for which the phonon properties are much better known than plutonium, as a test of method. In general, the experimental procedure was to lower the temperature to about 15K, measure the transmitted neutron intensity for successive intervals of 5 minutes for a total time of a few hours, raise the temperature and repeat the procedure, concluding with a run near room temperature. Ratios were calculated between the 5 minute runs at given temperature, the short runs being rejected if inconstancy as a function of neutron energy indicated that temperature equilibrium may not have been achieved. In this process almost half the data for each temperature were rejected.

Detailed computer programs (suitable for a fast PC) were developed to analyze the data. The data were fitted in observed count form using a least squares method with the option of added simple Bayesian constraints. The centre-of-mass neutron cross section (σ_t in eq.1) was assumed to have the single level Breit-Wigner form in the region of a resonance (see eq.A.1 in the Appendix), with the option of interfering many-level correction terms for the effects of other resonances of the same isotope, and background cross section terms from other isotopes or elements. The resonance component of the cross section was convoluted with the Doppler broadening function according to the formalism of ref.[3] to give the effective cross section $\sigma_{\text{eff},t}$ of eq.1. The transmission of the neutron beam as a function of time-of-flight was calculated from these cross sections.

The various complicated resolution effects described above were folded into the transmission function, which was then multiplied by the open beam function and added to the background to give a final function to match the data.

1. Tungsten

Natural tungsten contains several isotopes. For the purpose of determining phonon moments, the 4.15eV resonance in the cross section of ^{182}W is favoured. For the measurements, pure metal foils of thickness 0.05mm (3.22×10^{20} atom.cm $^{-2}$) and 0.35mm (2.25×10^{21} atom.cm $^{-2}$) were used to best match the cross section expected over the peak and well into the wings of this resonance. Data were taken at 15K, 40K, 80K, 170K, 200K and 310K.

In the least squares fitting the neutron resonance parameters were found to vary little from their compilation values listed by Mughabghab et al [5]. Typical fits to the data are shown in Figs.1 and 2. The first Boltzmann-weighted moment as determined for the various temperatures are listed in Table 1. They are seen to be a little higher than the values calculated from a Debye model of the spectrum with the Debye temperature deduced from specific heat data [7]. The statistical error associated with the Doppler moments is ~ 0.5 meV.

Figure 1. Data and fit for neutron transmission by the thin sample of tungsten at 15K.

Figure 2. Data and fit for neutron transmission by the thick sample of tungsten at 15K.

Table 1. Weighted first phonon moment $\langle h\nu \rangle_T$ determined from Doppler broadening of 4.15eV resonance of ^{182}W as a function of temperature. The quantity $\langle h\nu \rangle_D$ in the third row is the same average for a Debye model spectrum with the tungsten Debye temperature 390 K [6].

$T(\text{K})$	15	40	80	170	200	310
$\langle h\nu \rangle_T(\text{meV})$	25.5	26.3	27.4	41.3	39.3	60.9
$\langle h\nu \rangle_D(\text{meV})$	25.2	25.3	26.9	36.6	40.7	57.6

Attempts to fit the second moment resulted in values which were unphysical i.e. rather lower than the square mean phonon energy. This tendency of the data to favour a low value of the second moment may indicate that the phonon spectrum is dominated by a single narrow peak. It is more likely however that it is an artifact of an insufficiently accurate representation of the highly skewed resolution function of the flight path 5 facility. The second moment is mainly determined (at low temperatures) by the coefficient of the third Hermite polynomial in the Doppler broadening function (see eqs.2 and 5), an odd function and therefore very sensitive in its fitting to deficiencies in the resolution function employed in modelling the data.

2. Tantalum

The natural element tantalum consists almost entirely (99.988%) of the 181 isotope. The most suitable resonance for Doppler broadening studies is at 4.28eV. Only the data at 17K on samples of tantalum metal with thickness 0.0127mm ($7.038 \times 10^{-5} \text{ atom.b}^{-1}$) and 0.0381mm ($2.111 \times 10^{-4} \text{ atom.b}^{-1}$) have been analyzed to date. Results were: neutron width $\Gamma_n = 3.51 \pm 0.08 \text{ meV}$ (cf. $3.60 \pm 0.16 \text{ meV}$ in ref.[8]), radiation width $\Gamma_r = 51.3 \pm 0.2 \text{ meV}$ (cf. $53 \pm 4 \text{ meV}$ in ref.[5]), $\langle h\nu \rangle_T = 18.0 \pm 0.1 \text{ meV}$. Attempts to fit the second phonon moment again resulted in low values. The Debye temperature of tantalum from specific heat measurements is 259K [6]. The Debye phonon model with this Debye temperature gives $\langle h\nu \rangle_T = 16.75 \text{ meV}$. Typical fits are shown in Figs.3 and 4.

Figures 3 & 4. Data and fit for neutron transmission by the thin and thick sample of tantalum at 17K.

3. Plutonium

The only plutonium metal available to make a sample for the 1993 experiments had an isotopic content mainly Pu-239, 0.013% Pu-238, 5.56% Pu-240, 0.235% Pu-241, and 0.025% Pu-242. The most suitable resonances for Doppler broadening measurements are the 1.057eV resonance of ^{240}Pu and the 2.676eV resonance of ^{242}Pu . ^{239}Pu and ^{241}Pu are fissile, making the absorption widths of their resonances much greater than those of the even isotopes, while their cross section forms are greatly distorted from the single-level Breit-Wigner form. The ^{240}Pu content of the available material is sufficiently high to render it extremely difficult to obtain a thin enough sample for measurements across the central region of the very strong 1.057eV resonance. In the event the sample supplied to us was reported to be 0.2cm thick. From the neutron transmission measurements, especially over the wings of the 0.296eV ^{239}Pu and 1.057eV ^{240}Pu resonances, it appears that it is more than twice as thick as this ($n \approx 0.022 \text{ atom.b}^{-1}$). Furthermore it seems that the Pu-242 content is approximately 0.04% (from the strength of the transmission dip of the ^{240}Pu 2.67 eV resonance). Even though the Pu-242 thickness is thus almost 4 times the nominal amount, this is still an extremely thin sample of ^{242}Pu , and the amount of information we can expect to obtain from the neutron transmission data is correspondingly limited.

In particular, the details of the Doppler broadening in the wings of the resonance cannot be measured, and independent determinations of the resonance radiation width and the first moment cannot be made. In earlier measurements of the 2.67 eV resonance [8], the radiation width has been assessed as $\Gamma_r = 25 \pm 1.5 \text{ meV}$. We have therefore determined $\langle h\nu \rangle_T$ for specific values of the radiation width ranging from 24 to 27.5 meV. The results for an assumed radiation width of 24 meV are shown in Table 2. At the other extreme an assumed radiation width of 27.5 meV implies a much lower Debye temperature (about 116K) for the 15K and 25K data, but gives unphysical values of $\langle h\nu \rangle_T$, being lower than $2kT$, at the intermediate temperatures. Implications for a comparison with the powder diffraction data of Lawson et al [9] are also indicated in Table 2.

Table 2. Weighted first phonon moment $\langle h\nu \rangle_T$ determined from Doppler broadening of the 2.67eV resonance of ^{242}Pu as a function of temperature. The statistical error on these estimates is about 0.2 meV. The quantity $\langle h\nu \rangle_D$ in the third row is the same average for a Debye model spectrum with a Debye temperature of 174 K. The fourth row gives the inverse weighted phonon moment calculated with the same model, and for comparison the fifth row shows the same quantity derived from the Debye-Waller factors measured in a powder diffraction study of α -plutonium [9].

T(K)	15	25	35	50	75	200	300
$\langle h\nu \rangle_T(\text{meV})$	10.6	11.7	11.8	12.4	15.5	35.6	57.4
$\langle h\nu \rangle_D(\text{meV})$	11.3	11.5	12.0	13.2	16.2	35.8	52.6
$\langle (h\nu)^{-1} \rangle_T(\text{meV})$	105	114	126	150	197	470	696
$\langle (h\nu)^{-1} \rangle_{DW}(\text{meV})$	90	97	106	124	162	417	673

Figure 5. Data and fit for neutron transmission by the sample of plutonium at 15K.

4. High T_c super-conductivity

In 1993, at the suggestion of G.Kwei, data were taken by W.J.Trela and K.Meggers on a few samples in the lanthanum barium cuprate series of superconducting materials. Only one of these had a stoichiometric content of barium that rendered the material superconductive. The samples were too thin to allow Doppler broadening analysis of either the copper or barium resonances, but the data on the 72.2eV ^{139}La resonance did allow some inferences on the phonon spectra carried by the lanthanum atoms. It was established, by taking ratios of the data for different materials at the same temperature, that there was a substantial difference in the mean lanthanum phonon energy between the superconductive and non-superconductive materials. These results were reported by W J Trela at the meeting of the Materials Society in Boston, November 1994.

No further analysis of these data have been made to date. We have, however, designed a much thicker set of materials in the same series, that should allow results on phonon properties of barium and copper atoms to be extracted.

IV. Conclusions and recommendations

It has been established by theory and model simulation that the neutron resonance Doppler broadening phenomenon can be used at LANSCE for applications in scientific stockpile stewardship and other science-technology areas. One of the most significant possibilities is dynamic temperature measurement, in which sufficient data can be obtained from a single neutron pulse from LANSCE to measure a temperature to better than 10% accuracy over a time interval of a few micro-seconds. Another significant area is the extraction of information on phonon spectrum moments specifically related to single species of atoms in a complex material.

Most of the work described in this report has been devoted to the analysis of the resonance data obtained in the LANSCE runs of 1993. This was the

pioneering use of the LANSCE flight path 5 facility for this kind of research. In consequence, many problems have had to be uncovered, for example the form of the moderation time spreading from the moderator-reflector assembly at the neutron source, and the magnitude of long-life phosphor decay in the lithium glass neutron detector operating in current-mode. In spite of these difficulties it has been found possible to obtain rather precise values of the mean phonon energy for metals such as tungsten, tantalum and, despite a very inadequate sample, plutonium.

These 1993 data sets remain a rich mine of information for further analysis. Much of the remaining uncertainty in the details of the resolution function, especially its long tails, could be circumvented by analyzing ratios of data sets. For example, ratios over a resonance for the same sample but for different temperatures could allow extraction of the second and probably the third phonon moments with considerable confidence. Computer programs to model this method require development. A ratio method, for different samples at the same temperature, has already been used in a semi-quantitative way for analysis of data on high- T_c materials, with success, but in that work a very primitive form of the resolution function was used.

On the experimental side, the nature of the moderator time-spreading, presently understood only from Monte Carlo modelling, needs to be confirmed. This could be done by measuring resonances on a very short flight path (say 10m) using a detector operating in discrete event mode. Further investigation of the long-life phosphor decays of the neutron detector are required. More measurements of the open-beam neutron spectrum and the background function, especially their run-to-run variations, are needed. More confidence in thermometry is required; very accurately controlled and known sample temperatures would allow more detailed phonon information to be extracted from the runs at intermediate temperature (say, 100K to 200K), at which the first Boltzmann-weighted phonon moment actually becomes a series with leading term proportional to the temperature and first and second order terms are proportional to the second and fourth phonon moments.

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Appendix

Resonance neutron methods for dynamic temperature determination: some dopant elements and achievable accuracies

Experiments on dynamic temperature measurement using observation of Doppler broadening of neutron resonances in a single pulse of neutrons from the LANSCE source are presently underway. An important design issue is the choice of element as a doping material for the temperature determination. Elements with strong, narrow resonances in their neutron cross-sections are required. In this note, I consider a number of elements that are potentially useful in this respect. These, with their candidate resonances are listed as the heads of columns in Table A. In particular, I have calculated the possible accuracy of temperature measurement with a single LANSCE pulse for a range of temperatures and element thicknesses.

Temperature information is contained in the effective cross-section in the region of a narrow resonance level. The form of the effective cross section is described by eqs.1-7 in the main text. The leading term of eq.2 and the approximate form of the Doppler width parameter at high temperature, eq.7, are the relevant features for heating experiments. The second term on the r.h.s. of eq.7 is a small correction, which can be calculated well enough from simple information on the phonon spectrum, such as the Debye temperature from specific heat data. For these calculations the neutron cross section of a free nucleus at rest is taken to be the single-level Breit-Wigner formula,

$$\sigma_t(E) = \pi\lambda^2 \Gamma_n \Gamma / [(E - E_\lambda)^2 + \frac{1}{4}\Gamma^2] \quad (\text{A.1})$$

(plus much smaller potential scattering and resonance-potential interference terms) where $2\pi\lambda$ is the De Broglie wave length for the motion of neutron and target nucleus in the centre-of-mass system (and E is the energy of motion in that system), Γ_n is the partial width for the neutron formation process, Γ is the total width of the resonance and E_λ the resonance energy. The resonance parameters used in the calculations are taken from Mughabghab *et al* [5].

Neutron total cross sections are measured with greatest accuracy by the transmission method. A sample (of uniform thickness) of the material of interest is placed in a well-collimated neutron beam some distance before a neutron detector. The attenuation of the neutron beam caused by the sample at specific neutron energy E is characterized by the transmission $Y(E)$, the ratio of intensity of neutrons arriving at the detector with the sample in-beam to the intensity in open beam. The transmission is related to the effective total cross section (eq.1) by

$$Y(E) = \exp[-n\sigma_{\text{eff}}(E)] \quad (\text{A.2})$$

where n is the number density of the nuclear species per unit area of the sample perpendicular to the neutron beam.

Our procedure is to simulate experimental transmission data for a number of well-known, strong resonances in the cross sections of heavy nuclei, and then to perform a least-squares analysis of these to determine sensitivity to the

parameters of interest. The effective Doppler-broadened cross section is first calculated for a specific temperature, from the resonance parameters and an approximate or assumed Debye temperature for the metallic form of the element. These cross sections are then used to generate values of the transmission function for a few representative sample thicknesses at energy intervals of 10 meV. Based on assumed open beam neutron detection and background count levels a relative standard deviation is determined. The calculated standard deviation and numbers from a pseudo-random number chain are then used to generate a specific random deviation and hence a simulated "experimental" value of the transmission at each energy. We then perform a least squares analysis on these simulated data, fitting to the parameter $\langle h\nu \rangle_T$ with the specific objective of finding the variance on this quantity and hence the relative accuracy of its determination.

The following experimental conditions have been assumed for the purpose of calculating accuracies. The neutron flux through a sample approximately 6m from the source is about $6 \cdot 10^6 \text{ s}^{-1}\text{eV}^{-1}\text{cm}^{-2}$ at 1eV. This is based on the calculation by Russell *et al* [10] for 70 μA average proton current at the LANSCE target, but with a reduction by a factor of 0.8 as measured by Koehler [11]. For a pulse rate of 20 Hz, a sample of 1 cm^2 area and a ^6Li loaded glass scintillator neutron detector in current mode, with efficiency as described by Bowman *et al* [5] and placed so that it intercepts at least 75% of the neutrons passing through the sample, the detected neutron rates (in open beam) should be

$$2 \cdot 10^5 \text{ eV}^{-1} \text{ per pulse at 1 eV}$$

and

$$3 \cdot 10^4 \text{ eV}^{-1} \text{ per pulse at 5 eV.}$$

Instrumental energy resolution has not been taken into account in these calculations. With the inclusion of this, the measured quantity is a convolution of the transmission function Y (eq.A.2) with the instrumental resolution function. The integral of the quantity $(1-Y)$ over energy across the resonance (the "area" of the transmission dip) is independent of resolution. If $n\sigma_{\text{eff},0} \geq 1$ (where $\sigma_{\text{eff},0}$ is the peak cross section of the effective cross section) the area already contains the essential information on temperature. Therefore, if the width of the resolution function is not significantly greater than the larger of the natural or Doppler widths the accuracy of temperature measurement should not be significantly degraded.

A practical detector position on flight path 5 that will assure interception of most neutrons through a 1cm^2 sample (at 6m) is at 10m. The proton pulse width at LANSCE is $0.25\mu\text{s}$ and timing channels for work in the eV region are normally set at $0.5\mu\text{s}$ or less. Hence, the main contribution to experimental resolution is from the spread in neutron moderation time at the LANSCE target assembly. According to calculations by Russell (see ref.[6]) this spreading time, expressed as the standard deviation of an assumed gaussian function, is $2.8\mu\text{s}/E_n(\text{eV})^{0.394}$. The energy resolution for a 10m flight path resulting from this spread is given for each resonance entered in the Table below; it is given as $\sqrt{2}$ times the standard deviation to effect immediate comparison with the Doppler broadening width Δ and the resonance half-width $\Gamma/2$.

Table A should enable rapid decisions to be made on choice and thickness of dopant elements to be employed. On purely physics grounds, one choice stands out for almost every situation; this is the 1.06 eV resonance of ^{240}Pu . If an isotope of plutonium is ruled out on safety grounds, then the choice will depend on expected temperature rise in the dynamic experiment and the amount of dopant that can be tolerated. For the thinnest amount of material considered here, $n = 0.165 \times 10^{-4}$ atoms b^{-1} , and a modest temperature rise to, say, 600K, the 1.46 eV In resonance appears the best choice, but at higher temperatures the 3.91 eV Tm resonance becomes a little better. It should be noted, however, that the target nucleus for three candidate resonances, ^{152}Sm , ^{162}Dy and ^{178}Hf , are present to only about 25% in their natural elements. If the separated isotopes were to be used as the dopant material both ^{152}Sm and ^{178}Hf would yield higher accuracy than In for the thinnest amount of dopant. It should also be noted that most of the advantage of the low energy resonances such as ^{240}Pu and In comes from the higher neutron detection rate stemming both from the $1/E$ dependence of the neutron flux and the decreasing efficiency of the detector with increasing neutron energy. If the detector efficiency were to be improved, accuracy would be gained for the higher energy resonances.

Table I. Properties of possible neutron resonances for dynamic temperature measurement, and comparable achievable accuracies. The doppler broadening and resolution functions are assumed to be gaussian in form and the parameters given are $\sqrt{2}$ times the standard deviation. Resolution is calculated for the detector at 10m from the neutron source. The quantity n_d is the thickness of the doping element in atoms per 10^{16} .

Nucleus	¹¹⁵ In	¹⁵² Sm	¹⁶² Dy	¹⁶⁹ Tm	¹⁷⁸ Hf	¹⁸¹ Ta	¹⁹⁷ Au	²³⁴ U	²³⁶ U	²⁴⁰ Pu	²⁴² Pu
Resonance (eV)	1.46	8.05	5.44	3.91	7.78	4.28	4.91	5.16	5.45	1.06	2.68
Half-width (meV)	38	100	84	50	55	28	70	14	13	17	14
Resolution (meV)	17	112	72	50	107	55	64	68	72	12	33
<hr/>											
<i>T</i> = 300K											
Δ (meV)	37	76	60	50	69	51	52	49	50	22	35
Acc.(%) for:											
$n_d=0.165$	23	43	123	23	45	66	58	23	40	8.4	17
$n_d=0.33$	14	28	65	17	26	35	35	13	21	7.5	11
$n_d=0.66$	9.7	25	37	16	18	20	24	8.6	12	6.8	7.9
$n_d=0.99$	8.9	24	28	15	16	15	23	7.3	8.9	7.1	6.9
<hr/>											
<i>T</i> = 600K											
Δ (meV)	52	107	85	71	98	72	74	69	71	31	49
Acc.(%) for:											
$n_d=0.165$	18	29	85	17	35	59	38	26	49	6.2	16
$n_d=0.33$	10	19	45	12	19	30	22	14	26	5.1	9.3
$n_d=0.66$	6.9	16	25	9.9	12	16	15	8.8	14	4.2	6.2
$n_d=0.99$	5.8	14	19	8.9	11	12	13	7.1	10	4.0	5.3

Table continued.

Nucleus	¹¹⁵ In	¹⁵² Sm	¹⁶² Dy	¹⁶⁹ Tm	¹⁷⁸ Hf	¹⁸¹ Ta	¹⁹⁷ Au	²³⁴ U	²³⁶ U	²⁴⁰ Pu	²⁴² Pu
Resonance (eV)	1.46	8.05	5.44	3.91	7.78	4.28	4.91	5.16	5.45	1.06	2.68
Half-width (meV)	38	100	84	50	55	28	70	14	13	17	14
Resolution (meV)	17	112	72	50	107	55	64	68	72	12	33
<hr/>											
<i>T</i> = 1200K											
Δ (meV)	74	152	120	100	138	102	104	98	100	44	70
Acc.(%) for:											
$n_4=0.165$	15	23	66	13	31	58	20	28	52	4.9	17
$n_4=0.33$	8.2	14	34	8.4	17	30	16	15	27	3.7	9.3
$n_4=0.66$	5.1	10	19	6.6	10	16	10	8.6	14	2.9	5.7
$n_4=0.99$	4.2	9.7	14	5.8	8.4	11	8.9	6.6	10	2.6	4.6
<hr/>											
<i>T</i> = 2400K											
Δ (meV)	105	215	170	141	195	144	147	139	141	62	99
Acc.(%) for:											
$n_4=0.165$	13	19	57	11	30	57	24	30	56	4.2	18
$n_4=0.33$	7.1	11	29	6.6	16	29	13	16	29	2.9	9.3
$n_4=0.66$	4.2	7.9	16	4.9	9.1	15	7.9	8.8	15	2.2	5.3
$n_4=0.99$	3.3	7.1	11	4.2	7.1	11	6.6	6.4	11	1.8	4.2