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CONF-770688--1
S. L. Ruby

Prepared for
New Directions in Mössbauer Spectroscopy
Argonne, IL
June 10-11, 1977

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Symmetric Radiant State in Nuclear Resonant Bragg Scattering

S. L. Ruby

Argonne National Laboratory, Argonne, Illinois 60439*

The purpose of this paper is to describe a qualitative way to understand the temporal behaviour of nuclear resonant Bragg scattering. If you need quantitative detailed analysis, then use the careful work of Hammon and Trammell¹ who give scattering amplitudes from crystals for monochromatic photons, fold in the incoming wave packet frequency distribution, and Fourier-transform into the time domain. The present analysis is very crude, but is helpful in reaching an 'intuitive' understanding of a subject that has generated its share of confusion.

In the usual non-resonant scattering of x-rays from the regularly spaced electrons of a crystal, there has been little need to explicate the temporal behaviour. Such scattering is treated using the time-independent scattering amplitudes of the atom for precise frequencies. The atoms can be combined into lines, or planes, or crystals, and the corresponding amplitudes developed. Where the scattering amplitudes are rapidly changing functions of frequency, as is the case for nuclear scattering from a crystal, one anticipates delays of about the nuclear lifetime. Such delays of about 100 nanoseconds have been seen in both incoherent² and coherent^{3,4} scattering experiments.

The scattering theory has been carefully applied to the case of an enriched non-magnetic Fe crystal illuminated on the Bragg angle with nuclear resonant light, and it has been calculated that the reflectivity of the crystal remains large for perhaps fifty line widths.¹ In the near future, the explicit time behaviour will be calculated since experiments of this kind are in development. The broad reflectivity suggests an enhanced radiative line width, but it is not easy to give an intuitive interpretation of its origin. The power densities in the problem clearly rule out stimulated emission. Trammell⁵ first showed that the excess line width increases with the number of planes. Zaretski and Lomonosov⁶ introduced the idea of the nuclear exciton, whose decay is enhanced and directional. Kagan⁷ has noted that on Bragg the internal conversion decay channel is weakened as if α is decreasing, and calls this 'suppression of the inelastic channel'. We wish to show the intermediate state in Bragg scattering is a 'super-radiant' symmetric state,⁸ and thus explain its rapid decay.

Simplify the crystal to N_x , N_y , and N_p nuclei in the x, y, and z directions with spacings a, b, and d. Assume 100% enrichment, suppress polarization directions, and fix the nuclei rigidly - no thermal vibration. In addition keep the density of nuclei in the

* Work performed under the auspices of the USERDA, Division of Physical Research.

planes, and the number of planes, such that there is not appreciable absorption (Born approximation). To allow for time delay experiments, this light is chopped much shorter than the nuclear lifetime: the wave packet thus has an energy spread much wider than the nuclear width. However, for an approximate calculation, we suppress the detailed dependence on energy, and ψ refers to all frequencies in the wave packet. And the incident power is low, so that even a single excitation in the crystal is infrequent. Such an excitation, or intermediate wave function, can be written as

$$\psi_{\text{int}} = (N_x N_y N_p)^{-1/2} \sum_{\ell} \sum_m \sum_n e^{i \vec{k}_e \cdot \vec{r}_{\ell mn}} |e_{\ell mn} g_s\rangle \quad (1)$$

Clearly the relative phases of each ket merely represents the arrival time of the plane wave at the various positions. Otherwise each nucleus is treated alike. It is the single excitation, three dimensional version of Dicke's symmetric state. If the $r_{\ell mn}$ are random, as in a glass, the behaviour of ψ_{int} will be incoherent or random, despite the symmetry of (1). Note that even in a regular crystal, if the excited state were created by such localized effects as β -decay or Coulomb excitation, ψ_{int} would not be the appropriate description.

What is the radiative decay behaviour of ψ_{int} ? It will decay to a final state $|g_1 g_2 \dots g_n\rangle = |G\rangle$ with a photon $\vec{k}_f$. Putting $\langle G | O_r | e_{\ell mn} g_s \rangle = M_0$, the matrix element is easily obtained.

$$M = M_0 (N_x N_y N_p)^{-1/2} \sum_{\ell mn} e^{i(\vec{k}_i - \vec{k}_f) \cdot \vec{r}_{\ell mn}} \quad (2)$$

which is exactly the usual x-ray result. Following traditional paths now, let $\vec{k}_i = k(\hat{i} \cos \theta_i - \hat{k} \sin \theta_i)$, $\vec{k}_f = k(\hat{i} \cos \theta + \hat{j} \sin \theta \sin \psi + \hat{k} \sin \theta \cos \psi)$, $\vec{r} = \hat{i} \ell a + \hat{j} m b + \hat{k} n d$, and calculate $M^* M$.

$$M^* M(\theta, \psi) = \frac{M_0^2}{N_x N_y N_p} \sum_{\ell mn} \sum_{\ell' m' n'} e^{i(\vec{k}_i - \vec{k}_f) \cdot (\vec{r}_{\ell mn} - \vec{r}_{\ell' m' n'})} \quad (3)$$

which leads to

$$M^*M(\theta, \psi) = M_o^2 \left[1 + \frac{1}{N_x N_y N_p} A \cdot B \cdot C \right] \quad (4)$$

$$\text{where } A = \frac{\sin^2 N_x \alpha}{\sin^2 \alpha} - N_x \quad \text{with } \alpha = \frac{ka}{2}(\cos \theta_1 - \cos \theta)$$

$$B = \frac{\sin^2 N_y \beta}{\sin^2 \beta} - N_y \quad \text{with } \beta = \frac{kb}{2}(-\sin \theta \sin \psi)$$

$$C = \frac{\sin^2 N_p \gamma}{\sin^2 \gamma} - N_p \quad \text{with } \gamma = \frac{kd}{2}(\sin \theta_1 + \sin \theta \cos \psi)$$

It is easily shown that M^*M has only two narrow exit channels - the transmitted and reflected beam. If eq. (1) had a random phase in each term, as is appropriate for β -decay, then M^*M would be independent of direction. By integrating the intensity M^*M over all directions, the radiative line width γ_r is found.

$$\text{Note that } \int_0^\infty d\epsilon \frac{\sin^2 N_{ze}}{\sin^2 ze} = \frac{\pi}{2} \frac{N}{z}. \quad \text{Also the result is specialized}$$

to non-zero values of θ_B .

$$\gamma_r = \gamma_o \left[1 + \frac{2\pi}{k^2_{ab} \sin \theta_B} N_p \right] \quad (5)$$

This shows that the intermediate Bragg state decays with an enhanced radiative width, or shorter lifetime, compared to an isolated excited nucleus.

For ^{57}Fe using $\theta_B = 45^\circ$, $\gamma_r = \gamma_o(1 + .03 N_p)$. The observed lifetime includes the unchanged incoherent internal conversion process whose coefficient $\alpha = 8.4$, and therefore the overall width γ corresponds to $\gamma = \gamma_o(1 + .03 N_p) + 8.4 \gamma_o = \gamma_o(9.4 + .03 N_p)$. For $N_p \approx 10^3$, one expects $T \approx 25$ nanoseconds, not the ordinary 100. Clearly, the 30-fold decrease in the radiative lifetime, from 1000 nsec to about 30 nsec, greatly reduces the fraction of times that the intermediate state can decay via the 110 nsec internal conversion channel.

It has been shown above, by direct calculation, that a Bragg intermediate state (ψ_{int} with special $\vec{r}$) decays differently than the more usual 'random' intermediate states. The differences are two sharp Bragg peaks in direction versus isotropic decay, and decreased lifetime. Can this surprising fact be made even simpler? Independently both Dicke⁸ & Trammell⁵ have discussed a two body system, each of which can be in the ground (g) or excited (e) internal state. They are labeled 1 and 2 on the basis of position, even though $ka \ll 1$ with k the reduced wavelength and a the separation distance. Now kets like $e^{i\phi_1} |e_1 g_2\rangle$ or $e^{i\phi_2} |g_1 e_2\rangle$ describe situations where a particular one of the atoms is excited. To describe that only one nucleus is excited, but not knowing which, is done by

$$\psi_{\text{random}} = \frac{1}{\sqrt{2}} \left\{ e^{i\phi_1} |e_1 g_2\rangle + e^{i\phi_2} |g_1 e_2\rangle \right\}.$$

Till now the phases ϕ_1 and ϕ_2 have been random, but if the excitation has been such that there is a definite phase difference ϕ between particles 1 and 2, then one needs

$$\psi = 2^{-1/2} \cdot e^{i\phi_1} \left\{ |e_1 g_2\rangle + e^{i\phi} |g_1 e_2\rangle \right\}$$

and this state is most easily described as a linear combination of the symmetric

$$\psi_S = \frac{1}{\sqrt{2}} \left\{ |e_1 g_2\rangle + |g_1 e_2\rangle \right\}$$

and the anti-symmetric eigenvectors.

$$\psi_A = \frac{1}{\sqrt{2}} \left\{ |e_1 g_2\rangle - |g_1 e_2\rangle \right\}.$$

The important point that $\gamma_S = 2 \gamma_0$ while $\gamma_A = 0$, is easily shown.

$$\begin{aligned} \left[\gamma = \int d\Omega \left| 2^{-1/2} \left(|e_1 g_2\rangle \pm |g_1 e_2\rangle \right) \cdot \vec{O}_r \cdot \vec{G}_{kf} \right|^2 \right. \\ \left. = \int d\Omega \frac{M_0}{2} \left| 1 \pm 1 \right|^2 = \gamma_0 (1 \pm 1) \right]. \end{aligned}$$

Note that the random intermediate state can be described as a linear combination of ψ_S and ψ_A , and will have $\gamma = \gamma_0$. If this is generalized to n atoms instead of two, $\gamma_S = n \gamma_0$ while all the $(n - 1)$ anti-symmetric states have zero decay rates. If you can prepare symmetric states, they decay rapidly!! In electrical engineering language, to give the classical version, two close dipoles decay between four or zero times more rapidly than one, depending on the phase difference. So, at the end, the surprising (?) properties of enhanced, directional decay for nuclei become the old-fashioned properties of a phased array of antennas.

It is worth noting again that just having a high density of scatterers and an exciton is not enough for enhancement. For example, moving θ away from θ_B by a fraction of a degree will kill off the enhancement completely. Generally, enhanced decay rates are not found merely when more scatterers are used, for the

amplitudes add constructively in some regions of space, destructively in others, and overall the integrated intensity stays fixed. Only if the amplitudes add constructively every where in space will line widths increase.

Note that in the above description, the photon occupation number is zero when we calculate γ_r . The above symmetric radiant state has been specialized to only one excited nucleus. Dicke has considered super-radiant states with n atoms, of which m are excited ($m < n$). The temporal evolution of such states can involve stimulated emission, but not for our case.

Such states are not confined to γ -ray optics, of course. Analogous behaviour takes place in neutron scattering, and perhaps also with visible light.

A final 'intuitive' description - the incident plane wave interacts with many nuclei at once. Except at the Bragg angle, there is only incoherent summing of amplitudes, and the results are as if each nucleus scatters individually. At Bragg, the increased amplitudes for radiative scattering coming from the coherent symmetric addition leads to enhanced radiation widths for both absorption and reemission. Since internal conversion cannot be coherent with the final state marked by a K-hole, and is not speeded up, it is observed less frequently as T_{rad} decreases. It is more insightful to talk, not of suppression of the inelastic channel, but enhancement of the radiative channel.

I wish to acknowledge the tolerance of many colleagues as I tried to clarify these ideas. In particular, J. Hannon, M. Peshkin, P. Flinn, H. Lipkin, and R. Ringo all helped eliminate many incorrect earlier versions.

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