PHYSICAL REVIEW B VOLUME 49, NUMBER 2 1 JANUARY 1994-II

Time evolution of incoherent nuclear scattering from ^{57}Fe excited with synchrotron radiation

U. Bergmann,* J. B. Hastings, and D. P. Siddons
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973
(Received 27 October 1993)

We report the observation of the time evolution of incoherent resonant scattering from the 14.4-keV level of ^{57}Fe excited with synchrotron radiation. The measured time spectra are consistent with a model with two components. In this model, one component has a significantly enhanced decay rate and is the result of recoilfree excitation. The other component has the natural decay rate for an isolated nucleus and results from excitation with recoil.

In a pioneering experiment Seppi and Boehm[1] used broadband excitation to observe the nuclear fluorescence from ^{19}F and ^{55}Mn. In their conclusions they point to the future stating that the use of pulsed sources and time gated detectors would qualitatively improve such measurements. Synchrotron radiation (SR) is just such a source and has been the basis of a wide range of nuclear resonant scattering experiments (e.g., Refs. 2–14).

All of these experiments have concentrated on Mössbauer isotopes and in particular on the measurements of the coherent nuclear resonant scattering, such as nuclear Bragg scattering (NBS) and nuclear forward scattering (NFS). Both phenomena are directly connected to the Mössbauer effect and well understood theoretically, showing quantitative agreement with experiment.

To measure the coherent channels is an obvious choice for several reasons. First, for a typical sample thickness the intensity in those channels is much higher than in the incoherent channel and the scattered radiation is well collimated compared to the 4π sr emittance from incoherent scattering. Both facts make the detection of coherent scattering much easier. Therefore it is not surprising that an attempt to observe the incoherent channel performed in 1978 (Ref. 15) led to an ambiguous result. Second, NFS and NBS contain a variety of information in their time evolution, such as magnetic fields and electric field gradients, isomer shifts as well as polarization properties and strengths of hyperfine split transitions. Furthermore, their time evolution is dependent on phase relations between different scattering sites and different scattering geometries. All these properties are not only powerful in the investigation of solids containing Mössbauer isotopes, they also can be used to check basic physical principles.

Analogous arguments for the advantage of photoexcitation with SR resulting in incoherent scattering can be made. In particular the SR replaces the nuclear parent of conventional perturbed angular correlation (PAC) studies. It thus eliminates complications introduced by the large pertubations caused during the decay processes, e.g., change of chemical species and effects of decay products. Photoexcitation also does not require a three level system because the incident photon beam defines a reference direction and time for the excitation process. Finally as discussed below, the broad energy band of SR excites both the recoil-free and the recoil fraction eliminating the requirement for a large Lamb-Mössbauer factor as is the case for coherent scattering.

In this paper we report the measurement of the time evolution of incoherent scattering from an ^{57}Fe foil. Incoherent scattering is emitted into 4π sr and the incoherent decay involves several processes. The directly emitted photons have 14.4 keV while the radiative decay of the core hole following internal conversion yields predominantly Fe $K\alpha$ photons with 6.4 keV.[16] Since the intensity is weak compared to NFS, for example, we designed a detector to collect as much as possible of the incoherent signal. This detector, shown in Fig. 1, consists of a plastic scintillator viewed by two phototubes. The sample was placed at the center of the scintillator block in which a hole was machined to allow the passage of the incident and forward scattered beams. The rest of the detector system was similar to that used previously.[10] Also some adjustments in the electronics were made to optimize the efficiency not only for 14.4-keV photons but

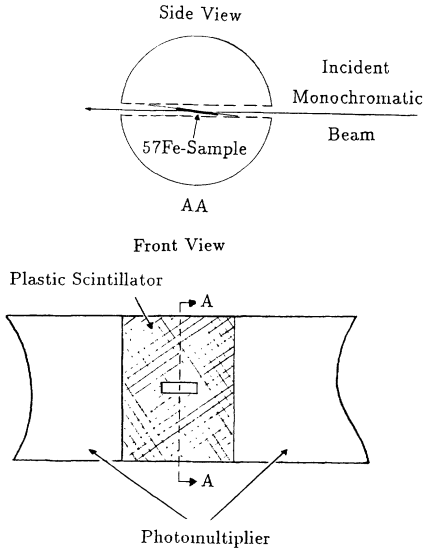

FIG. 1. Detector for incoherent scattering. The detector is shown from two views. The 0.5-μm thick ^{57}Fe foil is placed in the center of the scintillator block. By rotating the whole detector we obtained sample thicknesses of 1.9 and 3.1 μm depending on the angle between the incoming beam and the foil. The incoming beam had a bandwidth of approximately 10 meV after passing through our monochromator system.

also for the 6.4-keV Fe-$K\alpha$ photons. The sample was a 0.5-μm-thick highly enriched ^{57}Fe foil. By rotating the whole detector assembly the foil was inclined at two different angles towards the incoming beam to result in two different sample thicknesses.

The experiment was performed at the wiggler beamline X-25 at National Synchrotron Light Source (NSLS). The monochromator was a dispersive set of asymmetric-symmetric Si (840) channelcuts (see Ref. 14) downstream of the high heatload monochromator which is part of the beamline. This arrangement resulted in an intensity of order 10^7 Hz in an energy band of approximately 10 meV.

The measurements were made on and off resonance each for about 13 hours for the geometry with the thinner sample and 10.5 hours for the geometry with the thicker sample. This was done using a protocol which essentially eliminated the influence of the incident SR beam decay on the background subtraction. The off resonance measurement was done by tuning the monochromator 70 meV below resonance which is about 35 times the recoil energy and twice the energy corresponding to the Debye temperature of Fe. The delayed intensity in the time window between 25 and 200 ns was of order 0.5 Hz with background of order 3 Hz. The multi-channel analyzer channels were combined in 40-channel groups giving a time resolution of 6.5 ns per data point. The resulting point by point background subtracted spectra are shown in Fig. 2.

A possible spurious contribution to the observed signal which is important for thicker samples is due to x-ray fluorescence following NFS. The intensity from this process is emitted into 4π and depends on the efficiency of the absorber, in our case the ^{57}Fe sample. Since this process is simply an indirect detector of NFS, this contribution has the time evolution of NFS integrated from 0 to χ (χ is the effective sample thickness described below) but one has to include the quantum beats which occur in forward scattering from ^{57}Fe. In a seperate experiment we have observed this spurious effect. An additional ^{57}Fe foil (10 μm thick) was placed upstream of the incoherent scattering detector assembly such that only the forward scattered beam from this foil impinged on our sample. The measured intensity then includes both, forward scattering from the 10-μm foil followed by x-ray fluorescence showing quantum beats, and incoherent scattering from the sample inside the detector. We observed the quantum beats which were located at the same times as those from NFS of the 10-μm foil. This experiment shows a limit where such a contribution has to be considered. However our measurements of incoherent scattering for both chosen thicknesses do not show such quantum beats and we therefore neglect this contribution.

To describe the observed time spectra we had to include two contributions to the incoherent scattering. One results from excitation without recoil leading to an intensity contribution for incoherent scattering which we will call I_{RF} (RF stands for recoil free). The other, I_R, arises from excitation with recoil.

Let us first discuss the evaluation of I_{RF}. The probability for recoil free excitation is given by the Lamb-Mössbauer factor f which in the case of ^{57}Fe at room

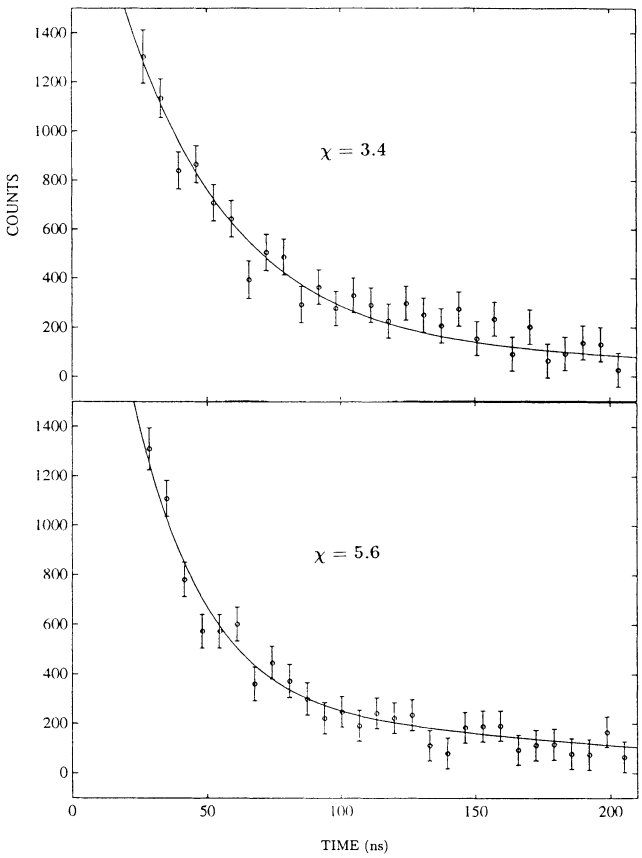

FIG. 2. Calculated intensity $I_{\mathrm{inc}}(t)$ compared with our experimental data for a foil with an effective thickness of $\chi = 3.4$ in the upper plot and $\chi = 5.6$ in the lower plot.

temperature is about 0.8 and therefore leads to a significant contribution. With this probability each nucleus in the path of the SR pulse participates in a collective excited state (exciton), in which a single excitation populates the ensemble of nuclei.[17] This exciton can decay through different channels. In each channel one observes a time evolution given by the total decay width. This decay width includes contributions from the coherent decay (proportional to the number of coherently excited nuclei) and the decay of an isolated nucleus either through a photon or a conversion electron. In addition interference effects (quantum beats) resulting from hyperfine splittings may occur. If the interference requires coherence between different nuclei (e.g., $\Delta m = 0$ transitions) it can only be observed in a coherent channel. If the interference is between excited levels leading to the same ground state, it can be observed in any channel.

In an incoherent decay process the atom at which the decay occurs changes its state and is therefore tagged. What is the decay rate for such a process? As mentioned above, it is given by the total decay width. Due to contributions of the coherent channel it depends on the number of nuclei involved. However, not all of the excited nuclei in the sample can contribute to the decay rate of the incoherent process. We will illustrate this important point with a gedanken experiment. Let us assume that the sample consists of many thin slices. In this case the exciton involves nuclei from all the slices. It is not affected by

any separation of the slices independent of how far they are separated, even though there can be a significant time difference for the arrival of the SR pulse. The reason for this is that the exciton is a phased state traveling with the speed of light, and no dispersion occurs by separating the foils. A measurement of the forward scattering would lead to identical results for arbitrary separations. However it is obvious that an incoherent scattering occuring in the first slice cannot be affected by, e.g., the last slice since the SR pulse might not have reached the last slice until after the deexcitation taking place in the first slice. Therefore in this case the nuclear exciton to be considered in the decay rate of the incoherent process cannot depend on the number of nuclei downstream of the tagged site. It is only determined by the number of excited nuclei upstream of the tagged site since they affect the wave impinging on the incoherent scatterer. If the slices are close together or not separated at all the incoherent decay rate is the same as in the gedanken experiment even though the deexcitation occurs mainly after the SR pulse traversed the entire sample. The reason for this is, that for an off-Bragg excitation (as we assume in our experiment) there is only one coherent wave in the forward direction and no coherent backscattering.

Therefore the decay rate observed from an incoherent scattering process at thickness χ' (see below) in the sample is the same as the NFS corresponding to this thickness. Interference effects in the excited state do not have to be considered if the incoherent signal is integrated over 4π sr, because the difference in the phases averages out those effects. However, there is a small contribution to the incoherent signal arising from diffuse elastic scattering associated with the "isotope effect." These effects are shown to be small.[18] The quality of our data does not allow the confirmation of any such contribution and therefore we neglect it. Thus, in our case, no interference term is included in $I_{\mathrm{RF}}(t)$. In our experimental setup we do not know at which location inside the sample the incoherent scattering occurs since the whole sample is inside the detector. We therefore have to consider each part of the sample equally. Replacing the sum of the individual contributions of each scatterer in the SR path by an integral over the sample thickness leads to the expression for I_{RF} given in Eq. (1):

$$I_{\mathrm{RF}}(\chi,\tau) \propto \int_0^{\chi} I_{\mathrm{NFS}}(\chi',\tau)d\chi' \tag{1}$$

where, neglecting any interference, the intensity for NFS called $I_{\mathrm{NFS}}(\chi',\tau)$ is given by[19,20]

$$I_{\mathrm{NFS}}(\chi,\tau) \propto e^{-\tau}\frac{\chi}{\tau}J_1^2(\sqrt{4\chi\tau}). \tag{2}$$

In these equations τ is the time in units of the natural lifetime Γ^{-1} (141.1 ns), χ is the effective thickness, and J_1 is the first order Bessel function. The effective thickness χ and the sample thickness d are related through the equation $\chi = d\mu_r/4$, where $\mu_r = 7.2\ \mu\mathrm{m}^{-1}$ is the nuclear absorption coefficient at resonance.

As a consequence, if the incoherently scattered photon is emitted from the upstream part of the sample, the decay rate is not much enhanced since the preceding collective excited state only includes a small number of nuclei. However, a photon emitted from the downstream end of the sample has an enhanced decay rate depending on χ. We have neglected electronic absorption in Eqs. (1) and (2). In Eq. (2) it only changes the scale factor and in Eq. (1) its contribution to the time evolution is negligible.

So far we have only considered incoherent scattering where the excitation is recoil free $I_{\mathrm{RF}}(\chi,\tau)$. A comparison of $I_{\mathrm{RF}}(\chi,\tau)$ and our measured data does not result in an acceptable agreement, especially for times greater than 60 nsec. We therefore included an additional contribution to the incoherent scattering.

In a conventional Mössbauer experiment the cross section for excitation with recoil is very small. The broadband SR pulse however, spans the energy changes due to recoil and therefore these excitations must also be included. The time evolution resulting from such a process shows a decay rate given by the natural lifetime Γ^{-1}. It is an inelastic process leading to an intensity emitted into 4π sr, either through a 14.4-keV photon or an internal conversion electron, which we detect by radiative decay of the resulting core hole. This contribution $I_R(\tau)$ can simply be written as

$$I_R(\tau) \propto e^{-\tau}. \tag{3}$$

Neglecting electronic absorption both contributions $I_R(\tau)$ and $I_{\mathrm{RF}}(\chi,\tau)$ should increase proportional to the sample thickness (number of nuclear scatterers). We will consider this by the proper normalization [see Eq. (4) and following]. We are not able to determine the branching ratio between $I_R(\tau)$ and $I_{\mathrm{RF}}(\chi,\tau)$. This is due to the fact that it was not possible in our experiment to know the detection efficiency for the different contributions to both $I_R(\tau)$ and $I_{\mathrm{RF}}(\chi,\tau)$. We therefore used a fit with three free parameters: the effective thickness χ, the scaling factor, and the branching ratio. To be consistent with our experimental setup we imposed the following two constraints on these parameters. First, since it was not possible to measure the absolute angle of the foil towards the incoming beam to better than 3°, we fitted χ with the constraint that the difference had to be consistent with the difference in angle. This angular difference between "thin" and "thick" could be measured to within 0.1°. Second, the branching ratio between $I_R(\tau)$ and $I_{\mathrm{RF}}(\chi,\tau)$ is assumed to be the same for both thicknesses since $I_R(\tau)$ and $I_{\mathrm{RF}}(\chi,\tau)$ increase proportional to sample thickness. The data were fitted to the intensity given by

$$I_{\mathrm{inc}}(\chi,\tau) = c[xI^*_{\mathrm{RF}}(\chi,\tau) + (1-x)I_R(\tau)] \tag{4}$$

where c is a scaling constant and x is the constant to give the branching ratio. $I^*_{\mathrm{RF}}(\chi,\tau)$ is normalized and defined as $I_{\mathrm{RF}}(\chi,\tau)/I_{\mathrm{RF}}(\chi,0)$. The normalization of $I_{\mathrm{RF}}(\chi,\tau)$ is necessary to keep the branching ratio between $I_{\mathrm{RF}}(\chi,\tau)$ and $I_R(\tau)$ constant for different χ's. Figure 2 shows the comparison of our measurements and the fit to $I_{\mathrm{inc}}(\chi,\tau)$. The solid lines in the figure correspond to $\chi = 3.4$ for the thinner sample and $\chi = 5.6$ for the thicker sample. This corresponds to foil angles of 15.3° and 9.3°, respectively. The scaling factor c was found to be proportional to χ in agreement with the prediction, and the branching ratio

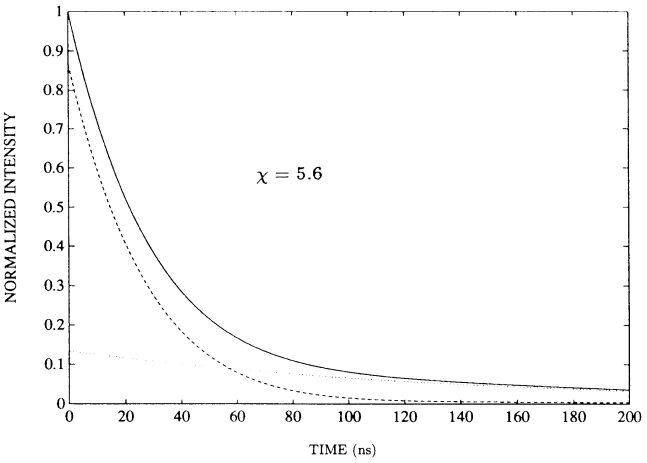

FIG. 3. Different contributions to the incoherent scattering. The contribution resulting from recoil free excitation $xI^{*}_{\mathrm{RF}}(t)$ is shown as a dashed line, $(1-x)I_R(t)$ (contribution resulting from excitation with recoil) is shown as a dotted line, and $I_{\mathrm{inc}}(t)$ is shown as a solid line. The effective thickness in this calculation is $\chi = 5.6$.

was found to be $x = 0.87$.

The important question is how well the data support the existence of $I_R(\tau)$. This can be answered by investigating the goodness of the fit, R, for models with and without this component. R is defined as

$$R = \frac{\sum_i \frac{(x_i^{\mathrm{exp}} - x_i^{\mathrm{calc}})^2}{\sigma_i^2}}{\sum_i \frac{(x_i^{\mathrm{exp}})^2}{\sigma_i^2}}, \tag{5}$$

where σ_i is the standard deviation, x_i^{calc} and x_i^{exp} are the calculated and measured data points. For our best fit, including $I_R(\tau)$, $R = 4\%$ and leads to $x = 0.87 \pm 0.03$, and $\chi = 3.4 \pm 0.34$ and 5.6 ± 0.56 for the thin and thick cases, respectively. Neglecting $I_R(\tau)$ ($x = 1$) leads to $R = 20\%$ with the sample thickness constrained to the known experimental error. Comparing this with our best fit demonstrates that including $I_R(\tau)$ in our model results in a significantly better agreement.

Although $x = 0.87$ has approximately the same value as the f factor for ^{57}Fe at room temperature, this may be fortuitous since as mentioned above we do not know the efficiency for detecting each component.

Figure 3 shows the contributions $I^{*}_{\mathrm{RF}}(\chi,\tau)$, $I_R(\tau)$ and $I_{\mathrm{inc}}(\chi,\tau)$ for the $\chi = 5.6$ fit illustrating that the contribution of $I_R(t)$ is significant at later times. Comparing Fig. 3 with Fig. 2 shows that $I_R(\tau)$ cannot be neglected. Based on this description the time evolution of incoherent scattering from samples with small or negligible f factors should be observable with SR. In fact it should be easier to observe than incoherent scattering from samples with large f factors.

In summary, we have observed the time evolution of the incoherent scattering from an ^{57}Fe sample. A simple model which contains two components is used to describe the measured time spectra. One is due to recoil free excitation and shows an enhanced decay rate, which is a function of sample thickness. Consistent with our data points at times greater than 60 ns is a 141-ns decay rate expected for isolated nuclei. This decay rate is expected for excitation with recoil and is therefore independent of the sample thickness. With the significant increase in intensity (more than three orders of magnitude) from undulator sources at next generation SR facilities angle resolved incoherent measurements (time dependent PAC) should be possible. The fact that our data are consistent with a model that includes the decay rate for an isolated nucleus demonstrates the potential of this technique for both Mössbauer and non-Mössbauer isotopes.

We especially thank Dr. L. E. Berman for his support at the wiggler beamline X-25 and Dr. P. M. Stefan and Dr. C. C. Kao for very helpful comments and discussions during and after the experiment. We also thank Dr. U. van Bürck for providing the very thin ^{57}Fe sample. Finally, the continued support of Professor J. P. Hannon is gratefully acknowledged. This work was supported by the U.S. Department of Energy under Contract No. DEAC02-76CH00016.

*Present address: Physics Department, State University of New York, Stony Brook, NY 11794-3800.

[1] E.J. Seppi and F. Boehm, Phys. Rev. **128**, 2334 (1962).
[2] E. Gerdau *et al.*, Phys. Rev. Lett. **54**, 835 (1985).
[3] E. Gerdau *et al.*, Phys. Rev. Lett. **57**, 1141 (1986).
[4] U. van Bürck *et al.*, Phys. Rev. Lett. **59**, 355 (1987).
[5] G. Faigel *et al.*, Phys. Rev. Lett. **58**, 2699 (1987).
[6] G. Faigel *et al.*, Phys. Rev. Lett. **61**, 2794 (1988).
[7] D.P. Siddons *et al.*, Phys. Rev. Lett. **62**, 1384 (1989).
[8] J. Arthur, G.S. Brown, and S.L. Ruby, Phys. Rev. Lett. **63**, 1629 (1989).
[9] H.D. Rüter *et al.*, Hyperfine Int. **58**, 2473 (1990).
[10] J.B. Hastings *et al.*, Phys. Rev. Lett. **66**, 770 (1991).
[11] R. Röhlsberger *et al.*, Europhys. Lett. **17**, 707 (1991).
[12] W. Sturhahn *et al.*, Europhys. Lett. **14**, 821 (1991).
[13] D.E. Brown *et al.*, Phys. Rev. Lett. **69**, 699 (1992).
[14] D.P. Siddons, U. Bergmann, and J.B. Hastings, Phys. Rev. Lett. **70**, 359 (1993).
[15] R.L. Cohen, G.L. Miller, and K.W. West, Phys. Rev. Lett. **41**, 381 (1978).
[16] H. Frauenfelder, D.R.F. Cochran, D.E. Nagle, and R.D. Taylor, Nuovo Cimento **19**, 183 (1961).
[17] J.P. Hannon and G.T. Trammell, Physica B **159**, 161 (1989).
[18] J.P. Hannon (private communications).
[19] Yu. Kagan, A.M. Afanas'ev, and V.G. Kohn, Phys. Lett. **68A**, 339 (1978); J. Phys. C **12**, 615 (1979).
[20] J.P. Hannon (private communications).