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Nuclear forward scattering of synchrotron radiation

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The coherent nuclear reemission in the forward direction has been measured following excitation of the Mössbauer transitions in polycrystalline ^{57}Fe foils by synchrotron radiation. The excited resonances are sufficiently narrow in energy that the time dependence of their decay can be straightforwardly observed, and in particular the details of the collective response to single-photon excitation can be studied. The collective nuclear scattering conditions were varied directly over a large range by using foil thicknesses between 0.5 and 28.5 μm. The envelope of the time evolution showed significant changes with increasing foil thickness. For thin foils (up to 3 μm) the envelope was exponential in the observed time window, with a decay time decreasing with increasing foil thickness. For thicker foils an oscillatory modulation of the envelope and a significant shift in the phase of the beat pattern were observed. A simple optical model can explain all the observed phenomena.

INTRODUCTION

After an initial proposal,[1] several attempts were undertaken to observe the relatively slow decay of resonant nuclei following excitation by the short pulse of synchrotron radiation (SR) (e.g., Ref. 2). However, the broadband nature of the transmitted or scattered SR means that the ratio of resonant to nonresonant radiation is extremely small (of order 10^{-9}). Such attempts met with little success until the technique of pure nuclear Bragg reflection was applied.[3] This technique relies on the properties of special single crystals containing the resonant nuclei to selectively diffract only the resonant photons from the broadband incident spectrum. These crystals are typically antiferromagnets or other similar structures which rely on magnetism and/or electric field gradients to modify the crystal symmetry for resonant photons. Although very successful in allowing the observation of SR-stimulated resonant Bragg scattering (see, e.g., Ref. 4 and references therein), the stringent sample requirements have so far limited the applicability of such a technique to the study of gamma optics principles.

Recently, a monochromator based on silicon crystals was constructed that allowed the direct measurement of nuclear resonant Bragg scattering even in the presence of strong electronic scattering.[5] This monochromator was subsequently used to demonstrate that a general SR-based Mössbauer spectroscopy is possible by measuring the time dependence of the coherent nuclear forward scattering NFS.[6] Related experiments using a similar monochromator were reported recently.[7] The monochromator also allows the straightforward investigation of collective effects in nuclear scattering, and it is these phenomena that are the subject of this paper. In particular, we address the time dependence of the NFS as a function of sample thickness, i.e., the number of coherent scattering centers per unit area.

The collective nature of the coherent interaction of the radiation with the system of nuclei should lead to a type of superradiance as defined by Dicke.[8] In the case of nuclear resonant scattering of single photons with short wavelengths (comparable to the lattice parameter), the possibility of superradiant decay has previously been predicted for the spatially coherent modes, i.e., both for nuclear Bragg scattering[9] (NBS) and for NFS.[10] These theoretical ideas about the collective effects were further developed, resulting in a detailed theory of nuclear resonant dynamical diffraction. The theory could describe the scattering of both conventional Mössbauer radiation[11] and pulsed SR,[12] and several interesting predictions resulted. Many of these phenomena have been experimentally verified (for a review, see, e.g., Ref. 13). In particular, enhanced decay rates have been experimentally demonstrated using NBS after resonant excitation by pulsed SR,[14] and also in NFS using Mössbauer radiation together with fast-shutter techniques.[15] The aim of the experiments described here is to demonstrate some of the phenomena predicted theoretically for NFS of SR as a function of sample thickness.

EXPERIMENTAL SETUP

The experimental arrangement used for these measurements was the same as that used in a previous experiment.[6] It comprised a special high-resolution x-ray monochromator that delivered a beam of 14.413-keV photons having a bandwidth of only 5 meV. This beam passed through our sample foils, which were placed in a vertical magnetic field. This aligned the sample magnetization parallel to the magnetic vector of the incident beam, and resulted in the $\Delta m = 0$ transitions being the only excitable transitions. The coherent nuclear decay in the forward direction was observed by a fast scintillation counter and recorded on a multichannel analyzer via a time-to-pulse-height converter. The time reference was generated by a separate fast silicon *p-i-n* photodiode that detected the white synchrotron radiation directly. This combination gave an overall time resolution of around 1 ns. The iron foils were all enriched in ^{57}Fe to around 95%. They varied in effective thickness between 0.5 and 28.5 μm. The National Synchrotron Light Source (NSLS) storage ring was operated in a special mode to optimize the stored electron-beam current and lifetime, while leaving sufficient time between successive SR excitation pulses for observation of the nuclear decay. The fundamental time structure of the NSLS x-ray storage ring consists of 30 "bins" each spaced 18.9 ns apart, which may or may not be occupied by electron bunches. For these experiments only every fifth "bin" was filled, with the rest ideally completely empty. In practice, some of the stored electrons occasionally fell into time "bins" adjacent to the desired ones (around 10^{-5} of them). This gives rise to sharp peaks in the spectra that are irrelevant to the nuclear-decay process. Such peaks are labeled in the data to avoid confusion. Counting rates in the delayed time window between 15 and 85 ns were around $1-2\ \mathrm{s}^{-1}$, for a typical stored electron current of 150 mA. The observed time spectra are shown in Fig. 1 (thin foils) and Fig. 2 (thick foils). No background corrections were made to the spectra.

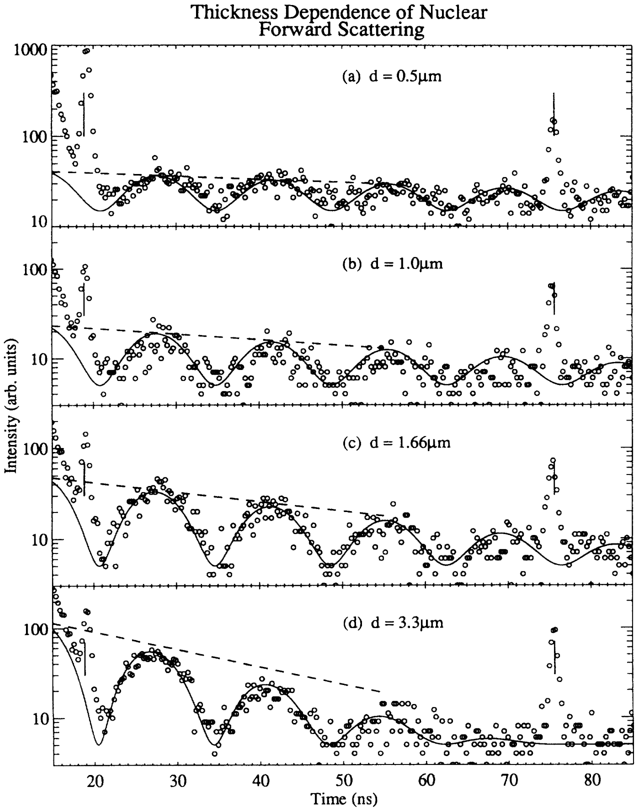

FIG. 1. Time spectra of the NFS from thin polycrystalline ^{57}Fe foils of different thickness. The foil thicknesses were (a) 0.5 (b) 1.0, (c) 1.66, and (d) 3.3 μm. All spectra were measured for 3000 s. The solid curves are calculations using Eq. (6) with parameters given in the text. The dashed lines indicate the exponential decay of the envelope as calculated using Eq. (4). The vertical bars at 18.9 and 75.6 ns indicate the location of the adjacent electron "bins" in the stored electron beam, which would ideally be empty. In fact, a small number of electrons do populate these "bins", giving rise to the sharp peaks observed.

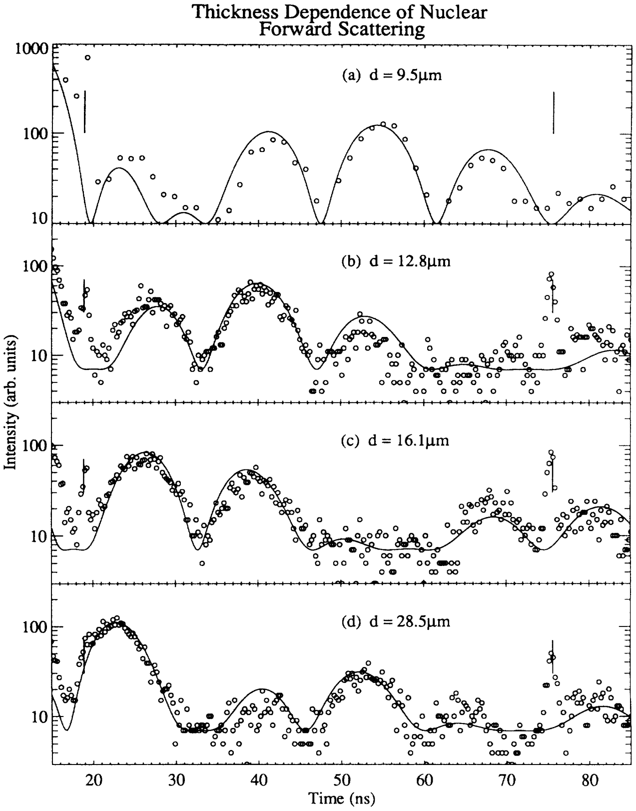

FIG. 2. Time spectra of the NFS from thick polycrystalline ^{57}Fe foils of different thickness (a) 9.5, (b) 12.8, (c) 16.1, and (d) 28.5 μm. All spectra were measured for 3000 s, except for (c), which was measured only for 2000 s. Spectrum (a) was measured during the single-bunch mode of the storage ring, where, due to the larger time window, the density of the data points is about 5 times less than in the other spectra. The remaining spectra were measured using the same five-bunch filling pattern as Fig. 1. As in Fig. 1, the vertical bars label the positions of unwanted electron bunches in the storage ring. The solid curves (a)–(c) were calculated using Eq. (6), while that in (d) required the use of a numerical Fourier-transform procedure to give acceptable agreement.

DISCUSSION

In both NFS and NBS, the excitation of the sample by a single photon is excitonlike; that is to say, it is not possible to localize the excitation to one particular nucleus. For the coherent decay of such a collective single excitation state, a strongly enhanced (superradiant) decay rate is predicted by the theory, where the enhancement increases with the number of the collectively acting scattering centers.[9,10,12,16,17]

NBS by crystals containing resonant nuclei involves a dynamical diffraction theory in which Maxwell's equations are solved in a triply periodic dielectric medium.[11] Near a Bragg reflection appropriate solutions involve two waves propagating coherently through the crystal. These waves interfere and produce a standing wave inside the crystal. Under appropriate conditions this standing wave can reduce or even eliminate the resonant absorption.

This is referred to in the literature as suppression of the inelastic channels[11] and is closely related to the x-ray Borrmann effect (e.g., Ref. 18). The nuclear dielectric constants are complex tensors involving the different resonance lines and their polarization properties.

In NFS, by contrast, only one wave has significant amplitude, and hence no suppression of absorption is possible. The dynamical theory of scattering under these conditions is familiar in optics and leads to the concept of the refractive index.[19] The nuclear refractive index is also a tensor, and its imaginary components are connected to the absorption cross section through the optical theorem. Its real components, required by causality, give rise to phase shifts in the transmitted waves.[20] This leads to the usual Kramers-Kronig relationship between the real and imaginary components of refractive index. This formalism can be applied to any resonant-scattering phenomenon, whether arising from photon-electron, photon-nucleus, or neutron-nucleus interactions. The outstanding feature of the ^{57}Fe Mössbauer resonance is the very small energy width and the consequent long lifetime of the excited state. Thus, a coherent interaction between a resonant photon and an ensemble of nuclei involves a phase memory of the excited state of more than 100 ns, i.e., 10^{11} oscillations. This is one of the few examples where the time-domain behavior can be directly observed. Electronic phenomena, in contrast, have lifetimes 10^6-10^9 times faster. In nuclear resonance scattering, one can therefore demonstrate the interrelationships arising from causality, and illustrate clearly the Fourier relationship between time and energy.

Experiments that attempt to elucidate the dependence of the collective effects on the number of scatterers using NBS are limited by crystal defects and by the requirement for very well-collimated sources and detectors. Forward scattering, on the other hand, makes no special demands on the sample. Consequently, using NFS, a detailed investigation of these collective effects is straightforward.

The time dependence of the forward-scattered radiation is the Fourier transform of the frequency-dependent amplitude spectrum of the transmitted wave. For a single isolated resonance line, this transform can be performed analytically. This has been done previously for the case where the incident beam is derived from a radioactive source,[21] and for the case of an incident SR pulse.[12,22] Classically speaking, one can say that in the first case the incident wave is extended in time, and so the decay takes place in the presence of the exciting wave. In the SR case, the resonant system is excited by a transient and left to decay freely.

If the selection rules allow more than one line to be excited, then the radioactive source can only excite one line at a time. The broadband SR pulse, on the other hand, excites all allowed transitions coherently, so that interference among them can be observed (quantum beats).[23]

ANALYSIS

Let us first consider in detail the response of a single isolated resonance. For a $\Delta m=0$ transition, eigenpolarizations are orthogonal linear polarizations, i.e., conventional σ- and π- polarizations. If our incident beam is essentially pure σ- polarized, the refractive index can be written as a complex scalar:

$$n(\omega)=1+\frac{\mu_r}{4k_0}\frac{\Gamma}{\hbar(\omega_0-\omega)+i\Gamma/2} . \tag{1}$$

In this expression, μ_r is the nuclear absorption coefficient at resonance and k_0 is the vacuum wave vector of the radiation of frequency ω. Γ is the natural half-width and ω_0 the center frequency of the resonance. The amplitude of the wave transmitted by such a medium is

$$A'(\omega)=A(\omega)\exp[-in(\omega)k_0 d] , \tag{2}$$

where $A(\omega)$ is the incident-wave amplitude at frequency ω. The incident spectrum of the SR that we consider is continuous, with equal amplitudes at all frequencies, i.e., $A(\omega)=1$. In the time domain, such a spectrum corresponds to a δ function at $t=0$. The effect of the resonant medium is to extend the time response to $t>0$. The detailed behavior of this delayed intensity is given by the Fourier transform of $A'(\omega)$, and depends on d, the sample thickness. The solution of the Fourier transform is[12]

$$I(t)\propto e^{-\tau}\frac{\chi}{\tau}J_1^2(\sqrt{4\chi\tau}) , \tag{3}$$

where $\chi=d(\mu_r/4)$ is the thickness[24] in units of $4/\mu_r$, and τ is time in units of the lifetime of the nuclear excited state, τ_0.

When $\tau<1/(1+\chi)$, Eq. (3) can be approximated by

$$I(t)\propto\chi^2 e^{-(1+\chi)\tau} . \tag{4}$$

This expression contains an exponential with decay constant $1+\chi$, i.e., the effective decay is still exponential, but faster than the natural decay by an amount that depends on χ. Equivalently, one can say that the effective energy, width of the resonance depends on χ, becoming broader but still approximately Lorentzian in form as the sample gets thicker. The intensity is proportional to the square of the thickness, in accordance with kinematical scattering.

For larger values of χ, the time decay is only exponential at very short times. For longer times the decay takes on the oscillatory Bessel-function shape described by Eq. (3). In the energy domain we have a distortion of the line shape. An alternative picture has been developed involving off-Bragg excitation of all radiative normal modes in the polycrystalline foil. These modes have slightly different frequencies and widths, so that their coherent superposition gives rise to the observed Bessel-function interference (normal-mode beats).[17,25]

When two or more lines are excited simultaneously, one must include the interferences between the two lines properly. If we restrict ourselves to the two $\Delta m=0$ lines, then the calculation of the refractive index simply involves an additional resonance term having a different resonance energy,[22] i.e.,

$$n(\omega)=1+\frac{\mu_r}{4k_0}\sum_{j=1}^{2}\frac{\Gamma}{\hbar(\omega_j-\omega)+i\Gamma/2} . \tag{5}$$

In a thin foil, where the separation in energy between the lines is large compared to their effective width, the resulting decay pattern is well approximated by multiplying the single-line pattern by a sinusoid representing the quantum beat resulting from the two lines. This gives

$$I(t) \propto e^{-\tau} \frac{\chi}{\tau} J_1^2(\sqrt{4\chi\tau}) \cos^2 \left[\frac{\Omega}{2} \tau_0 \tau + \phi d \right] , \qquad (6)$$

where Ω is the frequency difference between the two lines and ϕd is an additional phase shift discussed later.

As the effective linewidth gets larger, this approximation fails due to the increasing effect of overlap between the tails of the two lines, causing the lines to become strongly asymmetric. This regime can be calculated by numerical computation of the Fourier transform of Eq. (2), using the $n(\omega)$ of Eq. (5). All the features of NFS discussed above have been observed in our experiment, and in the following section we briefly point out these features.

RESULTS

In all the analysis presented here, residual intensity resulting from the excitation by previous bunches is assumed to be negligible. Detector background is accounted for by adding a constant intensity to the calculation.

For the thin foils (Fig. 1), the increase decay constant predicted by Eq. (4) is indicated by a dashed straight-line envelope over the data. Within our time window, this approximation is clearly good for panels (a) and (b), but fails for the thickest one (d) as expected. Of course, a longer time window would eventually reveal deviations from the exponential law even for the thin foils. The 3.3-μm foil [(Fig. 1(d)] is at the limit where the first Bessel minimum in the envelope is almost visible. The solid line in the figures is obtained by modeling the data using the approximate form [Eq. (6)], and this clearly gives good agreement.

For the thicker foils (Fig. 2), the Bessel modulation is clear, and a significant displacement of the quantum beats to earlier times is observed, which increases with foil thickness. A similar shift of the quantum beats proportional to the crystal thickness was recently observed in NBS of SR in Laue geometry.[26] The shift can be interpreted as due to the change of the refractive index at each resonance line by the influence of all other lines.[26] In the present experiment only the two $\Delta m = 0$ transitions influence each other. The refractive index near the lower-energy transition is increased by the influence of the higher-energy transition, while that at the higher-energy transition is reduced by the influence of the lower-energy transition. The effect of this asymmetric change of the refractive index at the positions of the two lines can be accounted for by an additional phase ϕd in the argument of the $\cos^2$- factor in Eq. (6). From the fit of all spectra a value $\phi = 0.03\ \mu\text{m}^{-1}$ was obtained, corresponding to a beat shift of 0.133 ns/μm, in agreement with the value calculated from μ_r and the separation of the $\Delta m = 0$ lines. However, taking into account this interference via an additional phase factor is only an approximation, and for larger thickness [for example Fig. 2(d)] it is necessary to perform the Fourier transform numerically as described above.

It is clear from the discussion above that the time dependence of the collective decay is not simply the Fourier transform of the conventional Mössbauer spectrum. Since absorption is proportional to the imaginary part of the refractive index (which is symmetric about ω_0), its Fourier transform would be symmetric in time. In order not to violate the causality principle, the (antisymmetric) real part of the index must be included to produce the correct time evolution.

SUMMARY

The influence on NFS from ^{57}Fe of different sample thickness has been studied in detail. For thin foils, the dependence of the exponential envelope on sample thickness was clearly demonstrated. For thick foils additional thickness-dependent modulations and a significant shift in the phase of the beat pattern were observed. All spectra showed the now-familiar quantum beats. The observed effects can be quantitatively explained using an optical model. Since that model involves coherent multiple scattering resulting in a macroscopic complex refractive index, we conclude that the features we observe do indeed result from coherent interactions. In particular, we point out that these narrow Mössbauer resonances are unique in that the time response is slow enough to be easily observed. The comparative simplicity of this time-domain refractometry encourages us to pursue the goal of a practical tool for synchrotron-based Mössbauer spectroscopy.

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