Time-Dependent Polarization in Mössbauer Experiments with Synchrotron Radiation: Suppression of Electronic Scattering

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The resonant forward scattering of x rays from ^{57}Fe nuclei is strongly polarization dependent. The broad-band excitation provided by synchrotron radiation (SR) results in an interesting time-dependent polarization mixing, which we will discuss. Further we demonstrate that by selecting only the component of the transmitted radiation which has a 90° rotated plane of polarization, the nonresonant (nonrotated) transmitted intensity can be substantially reduced. This new technique will allow full utilization of new powerful SR sources currently under construction.

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Resonant nuclear scattering of synchrotron radiation (SR) is slow compared to the scattering by electrons and therefore it is in principal possible to discriminate the resonant signal by using a fast detector and measuring the time evolution of the scattering [1]. It is the unfavorable ratio of delayed resonant signal to prompt background which makes the experiment so difficult. The SR experiment described in [2] used a conventional crystal monochromator together with pure nuclear Bragg reflections from isotopically enriched perfect crystals to suppress the electronic scattering. Reducing the monochromator bandwidth from several eV to 5 meV made it possible to perform the time analog of a conventional Mössbauer experiment [3]. Nevertheless, even with high resolution premonochromators, the ratio of prompt to delayed signal is still typically 10^6. Thus, independent of the source intensity, one is restricted by existing detectors to delayed intensities of a few counts per second. This signal level is already achieved at current SR sources. Full utilization of the next generation of SR sources, already being commissioned, will be conditional on the solution of the suppression problem.

In this paper we introduce a new technique for Mössbauer spectroscopy, in which the simple geometry of nuclear forward scattering (NFS) is combined with polarization sensitive suppression of the prompt electronic scattering. The central idea is to suppress the nonresonant electronic scattering by placing the sample between two crossed polarizers. The 14.4 keV transition of ^{57}Fe shows a hyperfine splitting with six allowed transitions, which have different polarization properties [4]. To a very good approximation, only the resonant part of the transmitted radiation can have its polarization state modified. This is the basis of the experiment described below. The demonstration experiment which we describe was performed on a dipole source at the National Synchrotron Light Source (NSLS) and therefore involves some compromise in the polarization analyzer adopted. The experimental setup is shown in Fig. 1. The first optical element consisted of a dispersive pair of asymmetric-symmetric Si channel cut crystals using the (840) reflection which has a Bragg angle of 45.1°. The first and last diffracting surfaces were cut at an angle of 39° to the (840) planes, while the second and third were cut parallel to the planes. This geometry [5] provides high angular acceptance (1.2 arcsec compared to 0.4 arcsec for the symmetric case), good energy resolution (9 meV compared to 27 meV), and an essentially full suppression of the remaining π polarization of the incoming SR beam. A calculation of this suppression using dynamical theory yields a value of order 10^{-12}, but this value should be treated with caution since it neglects the influence of crystal imperfections and of multiple and non-Bragg scattering. The sample, a 0.475 μm thick, highly enriched (95%) ^{57}Fe foil, was set nearly parallel (θ=17°) to the beam giving an effective thickness of 1.6 μm. In this geometry a magnetic field along the surface of the foil in the horizontal plane provides a large component parallel to the incoming beam. A beryllium single crystal was used as an analyzer. Its mosaic spread (about 0.1 mrad) determines the horizontal beam acceptance. The (00.6) Bragg reflection of beryllium is at 46.01° and provides a significant suppression of the horizontally polarized component [6]. Since this crystal is slightly mosaic it is difficult to calculate an accurate value for its suppression. The detector was the same as used in previous experiments (see, e.g., [3]). The NSLS storage ring operated in five and single bunch mode, which provides a time window of about 80 nsec (five bunch mode) [7].

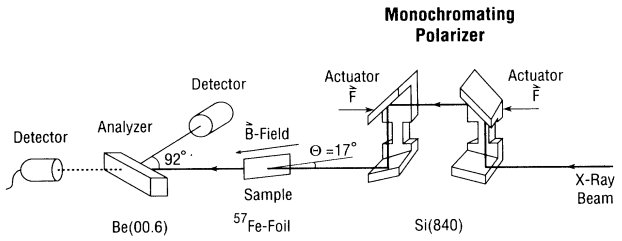


FIG. 1. Experimental setup.

Previous experiments were essentially classical Faraday effect measurements, using a single photon energy at any one time and observing a rotation of the plane of polarization which depended on the chosen photon energy and the sample thickness [8,9]. Experiments using SR are qualitatively different. The broad frequency band of SR excites all hyperfine split lines coherently and the rotation of the polarization is no longer thickness dependent. Instead the rotation becomes time dependent with a precession period depending only on the hyperfine splitting. For a σ-polarized incoming beam this leads to π- and σ-polarization components of the NFS with comparable intensities. Using crossed linear polarizers the σ component (prompt electronic and resonant scattering) can be almost completely suppressed, while the π component (only resonant scattering) is transmitted.

The theoretical treatment of the transmission of radiation through a Mössbauer absorber has been discussed by several authors [10–13]. The optical effects caused by resonant scattering can be described by considering a medium with a complex index of refraction expressed as a frequency-dependent 2×2 matrix $\tilde{n}(\omega)$. Since our polarizers are linear, we choose as a basis the linear polarizations π and σ, which are perpendicular and parallel, respectively, to the plane of the storage ring.

Consider the solution of the wave equation in a dispersive medium. The transmitted electromagnetic field can be written as

$$\mathbf{A}'(\omega) = \exp[i\tilde{n}(\omega)k_0 d]\mathbf{A}(\omega)\,. \tag{1}$$

In this expression d represents the effective sample thickness, k_0 the wave vector in vacuum, and $\tilde{n}(\omega)$ is given by

$$\tilde{n}(\omega) = 1 + \lambda^2 \rho \tilde{f}(\omega)\,, \tag{2}$$

where $2\pi\lambda$ is the wavelength in vacuum, $\tilde{f}(\omega)$ is the frequency-dependent 2×2 matrix representing the forward scattering amplitudes $f_{ij}(\omega)$ for a single nucleus, and ρ is the sample density. The subscripts ij refer to the polarization of scattered and incoming photons, respectively. The matrix elements $f_{ij}(\omega)$ depend on the orientation of the quantization axis with respect to the direction k_0 and polarization of the incoming x rays. They also depend on the nuclear transition amplitudes which contain the Lamb-Mössbauer coefficient, isotopic abundance, conversion coefficient, transition strengths, and hyperfine splittings. In general, each matrix element $f_{ij}(\omega)$ depends on all the nuclear transition amplitudes.

After substitution with Eq. (2), Eq. (1) can be factorized into a frequency-independent phase factor and the expression $\exp[i\tilde{f}(\omega)d]$ which represents the transmission operator. This operator can be evaluated by expressing $\tilde{f}(\omega)$ in terms of Pauli matrices. Expanding the exponential and using the algebraic rules of Pauli matrices gives the 2×2 transmission matrix $\tilde{T}(\omega)$ with elements $T_{ij}(\omega)$.

It is important to note that in general each element $T_{ij}(\omega)$ depends on all four matrix elements $f_{ij}(\omega)$ and therefore on all the nuclear parameters. Consequently, a measurement which is sensitive to only one of the $T_{ij}(\omega)$ is sufficient to probe all hyperfine levels.

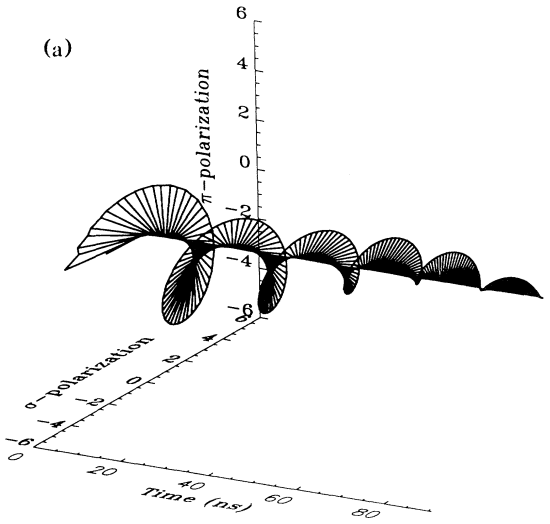
As discussed above, for an arbitrary orientation of the quantization axis with respect to the polarizer, the wave incident on the sample is not an eigenpolarization. For our choice of coordinates this means that $\tilde{T}(\omega)$ is not diagonal and all elements $T_{ij}(\omega)$ depend on all nuclear transition amplitudes. By specific choice of orientation one may emphasize the role of a subset of the manifold of transitions, for example the Faraday geometry reported here. In this case $\tilde{T}(\omega)$ is not diagonal and depends primarily on the $\Delta m = \pm 1$ transitions. The fact that $\tilde{T}(\omega)$ is not diagonal implies that polarization mixing occurs. It is this mixing of polarizations which we study in the experiment and use to suppress the electronic scattering. Although our measurements are performed in the Faraday geometry many other arrangements are possible, where all transitions contribute [14]. The only requirement is that the sample quantization axis can be oriented so that the incident polarization is not an eigenpolarization. We note that a different choice of basis set or coordinate system may result in a diagonal $\tilde{T}(\omega)$. In that case the matrices describing the polarizer and analyzer must be nondiagonal for polarization mixing to occur. That is, as stated above, the wave incident on the sample must not be an eigenpolarization.

We represent the polarizers by Jones matrices (see for example [15]). A given experiment is described by the matrix obtained by appropriate multiplication of $\tilde{T}(\omega)$ with the Jones matrices. The amplitude of the electromagnetic field at any position is represented by a vector to characterize its polarization state. In this convention σ polarization is given by the basis vector (1,0) and π polarization by the basis vector (0,1). Over the frequency range we are considering, the incident amplitude is constant (broad-band excitation).

Let us consider the situation with a polarized incident beam, where we detect the transmitted intensity without analyzer. The analog to Eq. (1) then becomes

$$\begin{aligned}\mathbf{A}'(\omega) &= \begin{pmatrix} T_{\sigma\sigma}(\omega) \\ T_{\pi\sigma}(\omega) \end{pmatrix} \\ &= \begin{pmatrix} T_{\sigma\sigma}(\omega) & T_{\sigma\pi}(\omega) \\ T_{\pi\sigma}(\omega) & T_{\pi\pi}(\omega) \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} A_\sigma \\ A_\pi \end{pmatrix}. \end{aligned} \tag{3}$$

The right-hand matrix represents the polarizer [our Si (840) monochromator] and for convenience we choose $A_\sigma = 1$ as mentioned before. A_π is small for SR and anyway unimportant if we consider the polarizer to be perfect. Our experimental conditions justify this approximation. The time-dependent amplitude $\mathbf{A}'(t)$ of the resonant scattering is related by a Fourier transformation to the frequency-dependent amplitude $\mathbf{A}'(\omega)$ from Eq. (3).



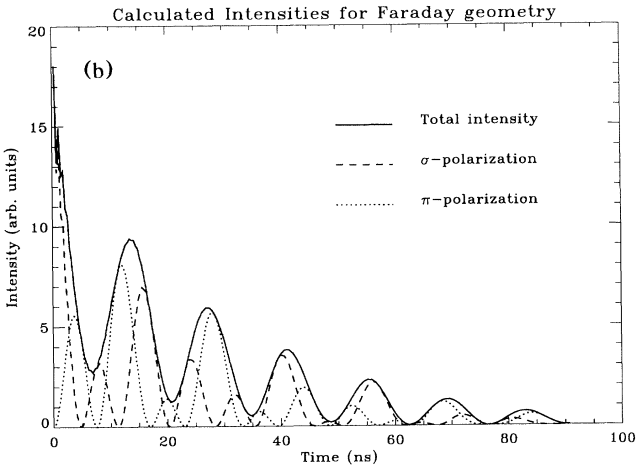


FIG. 2. (a) The calculated amplitude $\mathbf{A}'(t)$, in arbitrary units, of the transmitted wave and its polarization direction during the first 80 nsec after excitation. (b) The intensity components $I_{\sigma\sigma}(t)$ (dashed line), $I_{\pi\sigma}(t)$ (dotted line), and the total intensity $I(t) = I_{\sigma\sigma}(t) + I_{\pi\sigma}(t)$ (solid line), which is the square of the amplitude shown in (a).

Figure 2(a) shows this amplitude as a function of time and polarization direction. It can be seen that the radiation is always linearly polarized but that the plane of polarization is periodically rotating. Further, the amplitude is changing, reflecting the decay and beating of the collective excited state [16]. The intensity in our detector is simply proportional to the modulus squared of $\mathbf{A}'(t)$, i.e., $I(t) = I_{\sigma\sigma}(t) + I_{\pi\sigma}(t)$. This is shown as a solid line in Fig. 2(b). For our experimental conditions the splittings of the $\Delta m = +1$ and $\Delta m = -1$ lines are equal and lead to a single beat period of 14 nsec in $I(t)$. The figure also shows the $I_{\sigma\sigma}(t)$ (dashed) and $I_{\pi\sigma}(t)$ (dotted) components separately. They both show beating, but with twice the frequency of the total intensity beats.

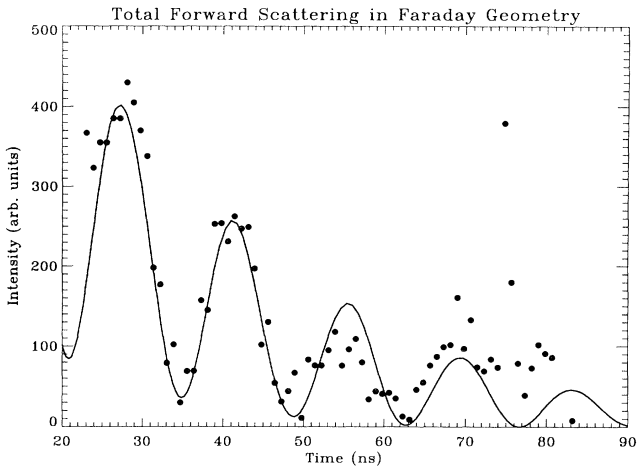


FIG. 3. Comparison between calculated and measured total intensity. No analyzer is used in this geometry.

This arises because the intensity of each component goes through a maximum (and a minimum) for both positive and negative amplitudes, i.e., twice for every rotation of the polarization vector.

Figure 3 shows the comparison between our measurement of $I(t)$ and the calculation shown in Fig. 2(b). The time-integrated intensity in the detector was 3.5×10^5 Hz, and the background subtracted delayed intensity in our time window (25–75 nsec) was about 0.5 Hz. This data clearly demonstrates the limitation of conventional SR forward scattering measurements. For the first 20 nsec the detector cannot recover from the prompt radiation burst. Further, any lack of purity in the electron bunch structure results in significant perturbations to the data as evidenced around 20 and 75 nsec in Fig. 3. It is important to note that this detection limitation is already severe using dipole radiation on a present day source. The use of undulator-based sources on existing and future machines will not result in the measurement of increased resonant intensities unless this problem is addressed.

To suppress the unrotated component before detection one can insert a crossed polarizer (analyzer) in the system and separate only $T_{\pi\sigma}(\omega)$, the rotated component. The matrix equation describing this is

$$\mathbf{A}'(\omega) = \begin{pmatrix} 0 \\ T_{\pi\sigma}(\omega) \end{pmatrix} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} T_{\sigma\sigma}(\omega) & T_{\sigma\pi}(\omega) \\ T_{\pi\sigma}(\omega) & T_{\pi\pi}(\omega) \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} A_\sigma \\ A_\pi \end{pmatrix}. \quad (4)$$

The left-hand matrix represents the beryllium analyzer, assuming that it is perfect. For electronic scattering the transmission matrix is essentially diagonal and application of Eq. (4) would lead to zero amplitude.

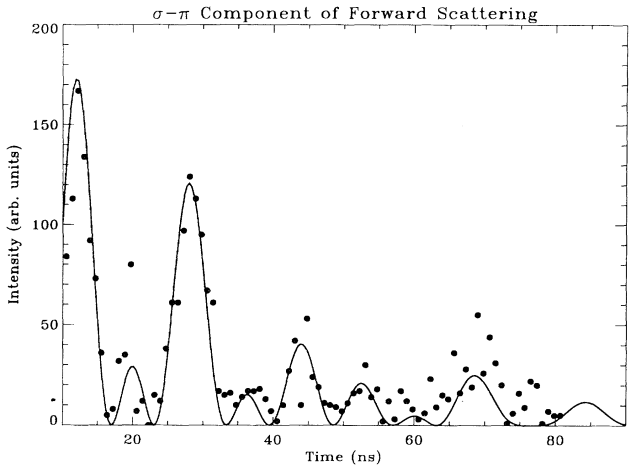


FIG. 4. Comparison between calculated and measured intensity of the rotated component $I_{\pi\sigma}(t)$.

Again the measured quantity is proportional to the modulus squared of $\mathbf{A}'(t)$ but now $\mathbf{A}'(\omega)$ is given by Eq. (4), i.e., $I(t) = I_{\pi\sigma}(t)$.

Figure 4 shows a comparison of the calculation with our measurement using the Be(00.6) analyzer to select only $I_{\pi\sigma}(t)$. The background subtracted delayed count rate was 0.05 Hz in the same time window. This rate does not include the first intense beat, which we can now observe because of our strong prompt suppression. The time-integrated intensity in the detector is only 80 Hz corresponding to a reduction of 4×10^3, while the resonant signal was only reduced by a factor 10.

In spite of the limitations of the Be(00.6) analyzer (Bragg angle limits on the extinction and reflectivity limits on intensity), a factor of 400 relative suppression has been achieved in these measurements.

In summary, we have observed the time-dependent nuclear Faraday effect in ^{57}Fe using SR. The results demonstrate that, by using the time-dependent polarization properties of NFS, the unwanted prompt scattering can be reduced by a large factor. In this work we show a suppression of 2 to 3 orders of magnitude. In a different context, polarimeter extinctions of order 10^6 have been demonstrated using perfect crystal optics [17]. This technology, which overcomes both limitations of the analyzer used in the present work, is optically well matched to the undulator sources of the next generation facilities.

Polarization mixing is not limited to the Faraday geometry. Given that the quantization axis can be properly oriented with respect to the incident beam, this technique can be applied if the magnitude of the relevant splitting is compatible with the experimental time window. The technique will completely remove detector-imposed limits on NFS intensities.

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