Switching Reciprocity On and Off in a Magneto-Optical X-Ray Scattering Experiment Using Nuclear Resonance of α-^{57}Fe Foils

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Reciprocity is when the scattering amplitude of wave propagation satisfies a symmetry property, connecting a scattering process with an appropriate reversed one. We report on an experiment using nuclear resonance scattering of synchrotron radiation, which demonstrates that magneto-optical materials do not necessarily violate reciprocity. The setting enables us to switch easily between reciprocity and its violation. In the latter case, the exhibited reciprocity violation is orders of magnitude larger than achieved by previous wave scattering experiments.

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The reciprocity principle, which states that the interchange of source and detector does not change the scattering amplitude of a wave scattering process, cannot be derived from first principles and is not necessarily fulfilled. The term "reciprocity" has been introduced by Stokes [1], and the numerous subsequent related publications cover the whole 20th century, as it is summarized in the review paper of Potton [2]. Reciprocity theorems were derived for various scattering problems, telling under which conditions and limitations the reciprocity principle is valid [3–7], and situations in the field of local and nonlocal electromagnetism [8–11], sound waves [12], electric circuits [13], radio communication [14], and local and nonlocal quantum mechanical scattering problems [5,15,16] were considered. Nonreciprocal devices (circulators and isolators) with on-chip integration possibility were also suggested [17]. In a recent work of Deák and Fülöp [18], a general reciprocity theorem was formulated, which covers all cases of wave phenomena that can be represented by a Schrödinger equation, with Hamiltonian $H = H_0 + V$, where H_0 describes free wave propagation and V the scatterer. Reciprocity is more general than time-reversal invariance, can occur for absorptive scattering media as well, and it also fundamentally differs from rotational invariance [18]. We note that, in x-ray optics, the term nonreciprocity may also refer to time-reversal odd optical activity [19–23], a meaning which differs from the historical one used here [2,18].

Here, we report on an experimental investigation of reciprocity and its violation, conceived based on the theoretical background established in Ref. [18]. Our aim was threefold: to show that magneto-optical materials do not necessarily violate reciprocity, to control easily whether reciprocity is present or missing, and to demonstrate that reciprocity violation can be a remarkably strong effect. The example considered belongs to the field of optics, where the multiple scattering of x rays or neutrons can be described by an index of refraction [24]. In forward scattering geometry, Blume and Kistner [25] established, already in 1968, the theory for Mössbauer absorption of γ radiation. The theory was later extended for grazing incidence scattering on stratified media [26–29] and computer programs also became available [30–34]. As a result, reciprocity situations can be simulated and the condition and aspects of reciprocity can be tested.

The experimental method is nuclear resonance scattering of synchrotron radiation that was introduced by Gerdau *et al.* [35], an analog of classical Mössbauer spectroscopy. The experiment was performed in forward-scattering geometry [36] corresponding to a conventional transmission Mössbauer arrangement. In contrast to laboratory Mössbauer experiments performed in the energy domain, nuclear resonance scattering experiments are performed in the time domain; i.e., the time response of the scatterer to the excitation by a synchrotron pulse is recorded and analyzed. The synchrotron radiation bunch excites the hyperfine-split nuclear energy levels simultaneously, leading to characteristic beats in the time spectra. The scatterer consisted of two foils mounted in a sample holder, each foil being a 6 μm thick ferromagnetic ^{57}Fe absorber uniformly magnetized in a field of 0.19 T of permanent magnets, as shown in Fig. 1(a).

We considered two experimental arrangements of the scatterer differing from each other in the directions of the magnetic fields acting on the iron foils according to Figs. 1(b) and 1(c) that we shall refer to, henceforth, as arrangements 1b and 1c, respectively. For both arrangements of the scatterer, the scattering time response (hereafter called "time spectrum") was compared to that of the reciprocal scattering. The interchange of source and detector position was realized, following [18], by a 180° rotation of the whole sample holder around the x axis of the coordinate system indicated in the figure.

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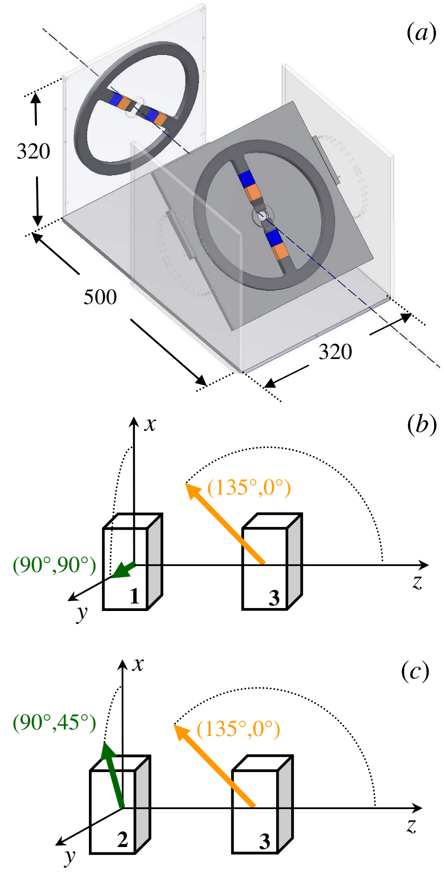

FIG. 1 (color online). (a) Sample holder for the reciprocity test of nuclear resonance scattering of synchrotron radiation (sizes in units of mm). The dashed line indicates the path of the synchrotron beam going through two slits of 0.5 mm diameter. The two α-^{57}Fe foils are mounted on the slits between permanent magnets providing 0.19 T magnetic field in the required direction, adjustable by the dark gray wheels mounted in the plane of the foils. (b),(c) Geometrical arrangement for the (magnitude) reciprocal and the nonreciprocal case, respectively. The synchrotron beam propagates in the direction of the z axis and gets scattered on foils 1, 3 (reciprocal scattering) or 2, 3 (nonreciprocal scattering). The thick arrows show the direction of the magnetic field in the iron foils, also given by the polar angles.

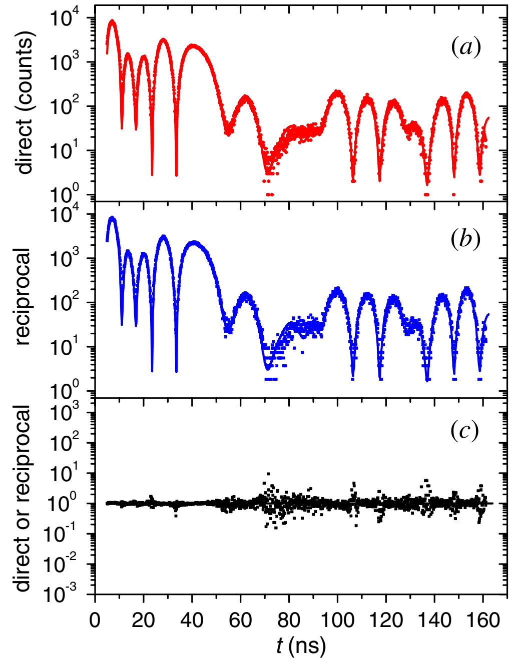

FIG. 2 (color online). (a) Measured (dotted lines) and simulated (solid lines) nuclear resonance forward scattering of synchrotron radiation time spectra on two α-^{57}Fe foils of 6 μm thickness using the polarizer-analyzer setup for incident σ (viz. horizontal electric) linearly polarized photons scattered to the same polarization ($\sigma \rightarrow \sigma$ scattering) for the case of hyperfine magnetic field orientations shown in Fig. 1(b) (foils 1, 3). (b) Results for the reciprocal (source-detector exchanged) situation realized by the 180° rotation of the scatterer [Fig. 1(a)]. (c) The amount of nonreciprocity, displayed as the ratio of the counts for the direct process (a) and of the reciprocal process (b). The ratio is found to be near the constant 1, which is the value when the intensities exhibit reciprocity. Slight deviations of the ratio from 1 can be seen in the regions of the minima of the beatings, where the count rates are low and statistical errors are significant.

The experiment was performed at the Nuclear Resonance side station ID22N of the European Synchrotron Radiation Facility (ESRF) delivering σ- (i.e., horizontally) polarized beam. The detector was a Si avalanche photodiode in front of which a Si(840) channel cut σ analyzer was applied. Since in this particular case both the source and the detector were of σ polarization, the reciprocal scattering could be realized by a mere 180° rotation of the scatterer. The four time spectra (count versus time diagrams) shown in Figs. 2 and 3 were measured with the respective arrangements 1b and 1c of the scatterer. Spectra (a) and (b) in each figure correspond to the direct and the reciprocal scattering geometry, respectively. Clearly, arrangement 1b provided an example where the measured intensity was reciprocal while arrangement 1c showed a large violation of reciprocity. How was it possible to find such scatterer arrangements?

In scalar wave phenomena, the interchange of source and detector defines the reciprocal process uniquely, but, for waves with more than one spin or polarization component, the polarizations of the reciprocal process must be chosen appropriately to obtain reciprocity. The conventionally used condition of reciprocity in linear systems [2] is the self-transpose (also called complex symmetric) property of the matrix of the scattering potential, of the index of refraction, of the dielectric or magnetic permeability tensors, or of Green's function [2,10,15]. This matrix must be considered in the polarization basis distinguished by the scattering processes in question [18]. For our scatterers, the potential V is zero in vacuum and, within each iron foil, is a space independent operator that depends on the direction (θ, ϕ) of the hyperfine magnetic field in the layer. The incoming synchrotron beam is σ polarized

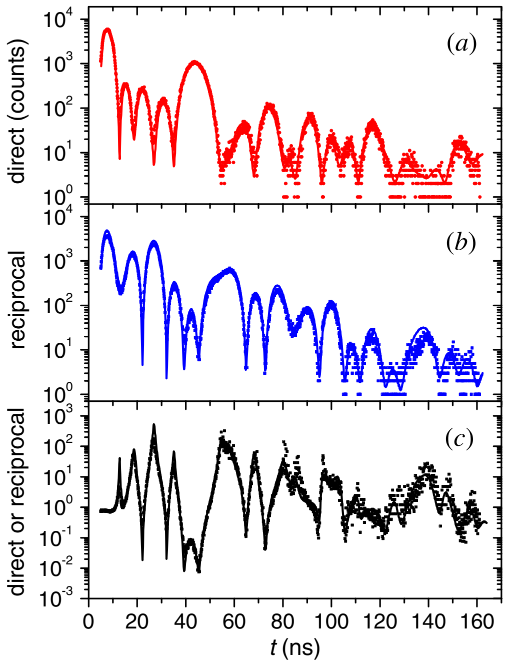

FIG. 3 (color online). Results for the hyperfine magnetic field orientations shown in Fig. 1(c) (foils 2, 3)—figure analogous to Fig. 2. Up to 3 orders of magnitude large nonreciprocity ratio is observed.

and the same polarization is measured by the analyzer, and it is the σ, π polarization basis in which self-transposeness of the scattering potential matrix V ensures reciprocity. For the $\Delta m = -1$ (Mössbauer) transition under discussion [37], this matrix reads

$$V = 2c\begin{pmatrix} 1-\sin^2\phi\sin^2\theta & -\frac{\sin^2\theta\sin 2\phi}{4}+i\cos\theta \\ -\frac{\sin^2\theta\sin 2\phi}{4}-i\cos\theta & 1-\cos^2\phi\sin^2\theta \end{pmatrix} \quad (1)$$

in a given layer with magnetic field direction (θ, ϕ), where the complex coefficient c is the same in both layers [18].

Let us consider three possible orientations for the in-layer magnetization, and the corresponding potentials,

$$(\theta_1, \phi_1) = (90°, 90°), \qquad V_1 = c\begin{pmatrix} 0 & 0 \\ 0 & 2 \end{pmatrix}, \quad (2)$$

$$(\theta_2, \phi_2) = (90°, 45°), \qquad V_2 = c\begin{pmatrix} 1 & -\frac{1}{2} \\ -\frac{1}{2} & 1 \end{pmatrix}, \quad (3)$$

$$(\theta_3, \phi_3) = (135°, 0°), \qquad V_3 = c\begin{pmatrix} 2 & -i\sqrt{2} \\ i\sqrt{2} & 1 \end{pmatrix}. \quad (4)$$

Apparently, V_1 and V_2 are self-transpose and V_3 is not.

A scatterer consisting of two layers with one foil having V_1 and the other having V_2 would mean a self-transpose potential and would obviously provide reciprocity. Therefore, so as to study nontrivial cases, in our experiment, arrangement 1b of the scatterer was a combination of a V_1 layer and a V_3 one, while arrangement 1c of the scatterer was formed by a V_2 foil and a V_3 one. Neither combination is self-transpose, so both situations are nonreciprocal.

The obtained experimental spectra can be seen in Figs. 2 and 3, respectively, together with the theoretical simulation predictions. We can see that the combination of V_1 and V_3 (i.e., arrangement 1b) results in reciprocity in the measured intensities, while the pair of foils with V_2 and V_3 (i.e., arrangement 1c) exhibits apparent nonreciprocity. The latter outcome is remarkable because of the large nonreciprocal effect: The ratio of intensities of direct and reciprocal scattering is nearly 10^3 in certain time intervals.

In parallel, the former result seems surprising since we have just found that reciprocity must be violated for that setting. The explanation is that, in experiments that detect probabilities or counts, i.e., only the squared absolute value of the complex scattering amplitude (i.e., intensity), reciprocity violation remains hidden if it is present only in the phase. Reference [18] calls such cases magnitude reciprocity and proves that, if there is a space independent angle δ such that

$$V_{12}(\mathbf{r}) = e^{i\delta}V_{21}(\mathbf{r}) \quad (5)$$

at any position $\mathbf{r}$, then magnitude reciprocity takes place. Applying this condition to our two scatterers, the first one (layers V_1 and V_3) proves to be a case of magnitude reciprocity—the phase factor "repairing" V_3 obviously "repairs" V_1 as well. Actually, our intention with this scatterer was to provide an example for the phenomenon of magnitude reciprocity, to drive attention to this weaker but important version of reciprocity. On the other side, "repairing" V_2 and V_3 requires two different phase factors. so there is no common phase factor for them.

The experimental results and the corresponding computer simulations made by the computer program [32] are both shown in Figs. 2 and 3. The slight imperfection in the agreement between measurement and simulation is due to, and informs about, nonperfect uniformness in the foil thickness and the magnetic field inside. The experiment kept these undesired influences under control by using the high collimation and brilliance of the synchrotron beam, which allowed the usage of slits of as small as 0.5 mm width selecting adequate homogeneous parts of both foils being 39.2 cm far from each others. The small size of the slits ensures that, after the 180° rotation of the sample holder, the same part of the foils is illuminated. The agreement of the experimental spectrum with that of the magnitude reciprocal counterpart setting—seen in Fig. 2—justifies the confidence that the recipocal situation was achieved to a high accuracy.

In summary, we have realized a (magnitude) reciprocal and a nonreciprocal experimental arrangement of magnetized α-^{57}Fe foils, which had neither time reversal invariance nor 180°-rotational symmetry. Using nuclear resonance scattering of synchrotron radiation, depending on the easily adjustable experimental geometry, reciprocity, and also 3 orders of magnitude large nonreciprocity, was experimentally observed in the intensities, in full agreement with the theoretical expectations. The presence of magneto-optic Faraday effect does not automatically lead to nonreciprocity. Further applications in the field of γ optics are expected, as nonreciprocal devices belong to an important class of optical components.

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