Large reciprocity violation switched on and off in a nuclear resonance scattering experiment

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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 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], and situations in the field of local and nonlocal electromagnetism [4, 5, 11], sound waves [12], electric circuits [13], radio communication [14], and local and nonlocal quantum mechanical scattering problems [3, 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 [23], 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 [23]. We note that, in X-ray optics, the term non-reciprocity may also refer to time-reversal odd optical activity [18, 22], which meaning differs from the historical one used here [2, 23].

Here, we report on an experimental investigation of reciprocity and its violation. 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 $\gamma$ 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 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 $\mu$m thick ferromagnetic $^{57}$Fe absorber uniformly magnetized in a field of 0.19 T of permanent magnets as shown in Fig. 1a.

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. 1b and 1c 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 [23], by a 180° rotation of the whole sample holder around the $x$ axis of the coordinate system indicated in the figure.

The experiment was performed at the Nuclear Resonance side-station ID22N of the European Synchrotron Radiation Facility (ESRF) delivering $\sigma$- (i.e., horizontally) polarized beam. The detector was a Si avalanche...
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 $\alpha$-$^{57}$Fe foils are mounted on the slits between permanent magnets providing 0.19 T magnetic field in the required direction, adjustable by the dark grey 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.

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 of 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
FIG. 2. (color online). (a) Measured (dotted lines) and simulated (solid lines) nuclear resonance forward scattering of synchrotron radiation time spectra on two $\alpha-^{57}$Fe foils of 6 $\mu$m thickness using the polarizer-analyser setup for incident $\sigma$ (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. 1b (foils 1, 3). (b) Results for the reciprocal (source-detector exchanged) situation realized by the 180° rotation of the scatterer (Fig. 1a). (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.

$V_2$ 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/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. Ref. 22 calls such cases magnitude reciprocity, and proves that, if there is a space independent angle $\delta$ such that

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

(5)

at any position $r$, 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 scaterer 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, 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 of 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 reciprocal situation was achieved to a high accuracy.

In summary, we have realized a (magnitude) reciprocal and a nonreciprocal experimental arrangement of magnetized $\alpha-^{57}$Fe foils, which had neither time reversal invar-
ance nor 180°-rotational symmetry. Using nuclear resonance scattering of synchrotron radiation, depending on the easily adjustable experimental geometry, reciprocity, and also three 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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37. We note that the measurement result contains Δm = 0 and Δm = +1 transitions as well but those cases will not require separate treatment concerning reciprocity, as the Δm = +1 potential is (proportional to) the transpose of the Δm = −1 one, and the Δm = 0 potential is automatically self transpose.