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Ultrafast inverse Faraday effect in a paramagnetic terbium gallium garnet crystal

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Conventional wisdom dictates that magneto-optical and optomagnetic phenomena are reciprocal and of equal strength. We test this assumption in a pump-probe experimental study of the ultrafast inverse Faraday effect in a terbium gallium garnet crystal. The thorough quantitative analysis of the observed polarization response unambiguously demonstrates a remarkable discrepancy of several orders of magnitude between the strengths of the direct and the inverse effects. This finding further questions the validity of standard magnetic models relying on the use of the static Verdet constant on subpicosecond time scales.

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I. INTRODUCTION

The development of femtosecond lasers has led to new prospects for the study of femtosecond condensed-matter phenomena.[1] Processes triggered in magnetic systems by optical pulses of femtosecond duration belong to the field of femtomagnetism. The observations of ultrafast demagnetization,[2] magnetization reversal,[3] and excitation of coherent magnons[4,5] have prompted both a theoretical scrutiny and the search for additional experimental evidence.[6] However, it has become clear that common models for magnetism and, more generally, condensed matter often fail to describe the highly nonequilibrium states observed on ultrashort time scales.

Important femtomagnetic phenomena are associated with the inverse Faraday effect (IFE). The IFE refers to the magnetization of media by circularly polarized light. The effect was predicted theoretically in 1961,[7] although some of its underpinning manifestations had been observed even earlier.[8] Pershan and co-workers reported phenomenological classical[9] and quantum[10] theories of the IFE as well as its experimental observation.[11] Perhaps the most important idea and conclusion of the papers were that the strengths of both the direct (conventional) and the inverse Faraday effects were described by the same parameter that characterized the magneto-optical properties of the medium—the Verdet constant V.

In this Rapid Communication, we report measurements of the strengths of both the direct and the inverse Faraday effects in a terbium gallium garnet (TGG) crystal within the same pump-probe experiment. We demonstrate a remarkable discrepancy of several orders of magnitude between the strengths of the direct and inverse effects, thereby challenging the common understanding of their relationship. In particular, our experiments emphasize the limitations of the use of the light-induced effective magnetic field in treating the optomagnetic phenomena in the ultrafast regime.

II. THEORY

Let us consider a linearly polarized electromagnetic plane wave propagating through a magneto-optical crystal, e.g., TGG. In applied magnetic field (magnetic-flux density) **B**, the crystal becomes gyrotropic, causing the polarization plane of light propagating in the crystal to experience so-called Faraday rotation[12] by angle,

$$\Delta\theta = VlB, \tag{1}$$

where l is the length of the sample. Equation (1) assumes that the crystal does not have any spontaneous magnetization, e.g., it is a paramagnet, such as TGG at room temperature. In this case, two main mechanisms contribute to the Faraday effect.[13] One contribution is due to the diamagnetic response of the material (i.e., induction of microscopic circular currents creating a magnetic field opposing the external magnetic field) and is, therefore, always present. Conventionally, the Verdet constant V_d, referring to the diamagnetic Faraday effect, is positive. The other mechanism is related to the paramagnetic magnetization of the material (i.e., alignment of its already existing microscopic magnetic moments by the external field) and is, therefore, only possible in materials containing such magnetic moments. Thus, the sign of the paramagnetic contribution is opposite that of the diamagnetic one. The total Verdet constant is equal to the sum of the two terms: $V = V_d + V_p$, where $V_d > 0$ and $V_p < 0$. In paramagnetic magneto-optical crystals, the paramagnetic term V_p is expected to prevail, at least, in the static regime.

According to conventional understanding of the IFE, light of elliptical polarization induces magnetization in a material even if there is no magnetic field applied. In samples with isotropic magneto-optical properties and insignificant absorption (e.g., in TGG), optically induced magnetization $\mathbf{M}^{\mathrm{NL}}$ can be written via complex electric field **E** of the incident light,[11]

$$\mathbf{M}^{\mathrm{NL}} = \chi_m^{(2)}[\mathbf{E} \times \mathbf{E}^*], \tag{2}$$

where $\chi_m^{(2)}$ is the nonlinear magneto-optical susceptibility. The phenomenological theory developed by Pershan[9] and Pershan *et al.*[10] for quasiequilibrium conditions states that $\chi_m^{(2)}$ is related to the Verdet constant,

$$V = -\frac{4\pi^2\chi_m^{(2)}}{n\lambda_0}, \tag{3}$$

where λ_0 is the wavelength of light and n is the corresponding refractive index. Then, the nonlinear magnetization due to light propagating along the z axis can be rewritten as

$$\mathbf{M}^{\mathrm{NL}} = \frac{\lambda_0 V}{2\pi c}(I_R - I_L)\mathbf{z}_0, \tag{4}$$

where I_R and I_L are the intensities of right and left circularly polarized light, respectively, and $\mathbf{z}_0$ is a unit vector along the z axis. In this picture, a circularly polarized optical pulse induces a pulse of nonlinear magnetization that follows

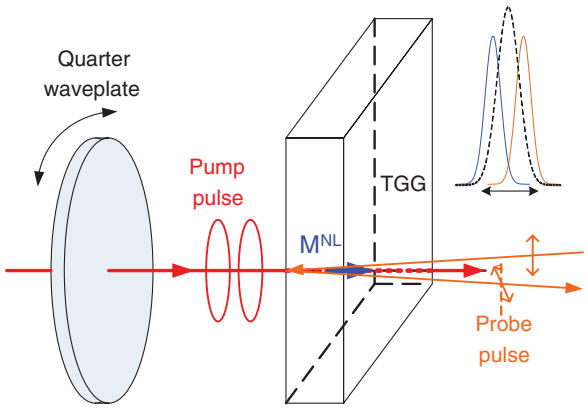

FIG. 1. (Color online) The experimental geometry is schematically shown. The TGG crystal is illuminated by the pump pulse normally incident from one side and the probe pulse incident from the opposite side at a small angle from the normal. The ellipticity of the pump beam is controlled by the $\lambda/4$ plate. The small angle between the pump and the probe beams allows us to separate them using an aperture. Varying the delay between the pump and the probe, we record the evolution of the Faraday rotation signal, which has the form of a convolution between the temporal profiles of the probe pulse and the pump-induced perturbation of the refractive index as shown in the inset.

the temporal envelope and transverse profile of the optical intensity. Assuming an infinitely small lifetime of the induced magnetization, the magnetization pulse propagates with the group velocity of the optical pulse. The process is analogous to the electro-optical rectification via the inverse electro-optical effect in crystals, such as ZnTe.[14] Indeed, the IFE could be referred to as "magneto-optical rectification."

III. EXPERIMENT

The measurements are performed on a $10 \times 10 \times 1\text{-mm}^3$ single crystal of TGG cut along a $\langle 111 \rangle$ plane. A Ti:sapphire laser generates 800-nm pulses with durations of approximately 100 fs and a repetition rate of 1050 Hz. Each pulse is split into a pump pulse and a probe pulse. The pump and probe illuminate the sample from the opposite sides, allowing a small deviation from the normal incidence (Fig. 1) so that the beams are nearly parallel inside the sample. A beam profiler is used to determine the diameter of the pump beam at the sample position as 1.5 mm at half maximum. The diameter of the probe spot is approximately 100 μm. By measuring the average beam power and taking into account the repetition rate of the laser, we calculate fluences and peak intensities of the pump and probe for particular values of the pulse duration and transverse spot sizes. In our setup, the intensity of the pump beam is varied between 0.4 and 31 GW/cm^2 (corresponding to fluences between 40 μJ/cm^2 and 3.1 mJ/cm^2), whereas, the probe intensity ($\sim$0.5 GW/cm^2) is fixed. The incident pump and probe are elliptically and linearly polarized, respectively, with the ellipticity of the pump polarization controlled by a quarter-wave plate. The probe pulse reflected from the rare surface of the sample is directed into an optical bridge detector to measure the polarization rotation acquired upon transmission through the sample. When the pump and probe pulses inside the TGG crystal are overlapped in time and space, the pump-induced change in the refractive index results in rotation of the probe polarization. Varying the pump-probe time delay, we record the time evolution of the polarization signal, given by the convolution of the pump-induced transients and the probe pulse.

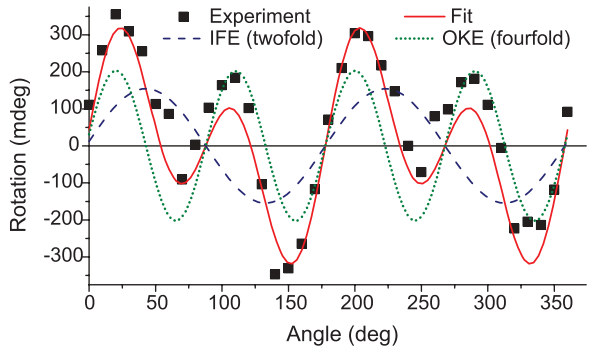

FIG. 2. (Color online) The measured rotation of the probe pulse polarization is shown as a function of the orientation of the quarter-wave plate, i.e., the helicity of the pump pulse (squares). The fitting curve (solid) shown on the graph consists of the twofold (dashed) and fourfold (dotted) sinusoidal contributions, which correspond to the IFE and OKE, respectively.

IV. RESULTS

In a cubic crystal, such as TGG, three different effects contribute to the nonlinear optical response: the IFE, the optical Kerr effect (OKE), and optically induced anisotropic polarization (OIAP).[15] Their different dependences on the pump and probe polarizations and the crystal orientation[15–17] allow us to isolate and to quantify the IFE's contribution. Indeed, the OIAP effect is maximized when the pump and probe are both linearly polarized parallel to certain orientations relative to the crystallographic axes. The IFE and OKE are absent in this case. There are also orientations of the crystal at which the OIAP signal is zero.[15] At such an orientation of the crystal, we isolate the IFE by setting the pump polarization to purely circular.[16,17]

Figure 2 shows the dependence of the signal amplitude upon the orientation of the fast axis of the quarter-wave plate relative to the polarization of the incident pump pulse. The dependence is fitted to the sum of twofold and fourfold periodic functions, corresponding to the contributions from the IFE and OKE, respectively.[16,17] The good fit confirms our assumptions about the origins of each contribution. Indeed, the IFE is maximized, whereas, the contribution from the OKE decreases to zero, when the pump is circularly polarized. Figure 3 shows typical rotation signals for the right and left circularly polarized pumps. The signal has a Gaussian-like shape and has opposite signs for the opposite pump helicities. Figure 4 presents the amplitude of the Faraday signal as a function of the pump intensity together with a linear fit. The linear intensity dependence indicates that the observed signal is a second-order nonlinear effect with respect to the electric-field amplitude of the pump.

The regular behavior of the measured signal allows us to quantify the contribution due to the IFE. The Verdet constant

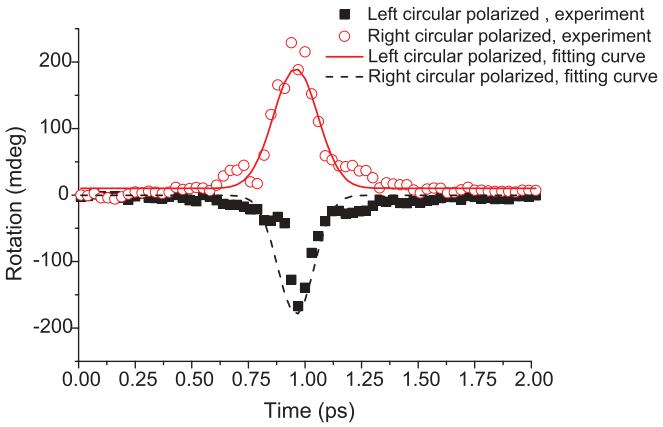

FIG. 3. (Color online) Measured rotation of the probe pulse polarization for the left (solid squares) and right (open circles) circularly polarized pumps. The fitting curves are Gaussian. The slight pedestal on both signals is due to the distorted temporal shape of the optical pulses.

of TGG at 800 nm is -0.28 min G^{-1} cm^{-1} [18] or -0.8×10^{-5} rad G^{-1} cm^{-1}. Using Eq. (4), we estimate the nonlinear magnetization induced by the pump pulse with a peak intensity of 30 GW/cm^2 (a fluence of 3 mJ/cm^2) as $M^{\mathrm{NL}} \approx 10^{-3}$ G. This magnetization is equivalent to that induced in TGG by applying an external magnetic field of

$$B_{\mathrm{eff}} = (1 + 4\pi\chi)M^{\mathrm{NL}}/\chi, \tag{5}$$

where χ is the linear magnetic susceptibility of TGG. Using the room-temperature magnetic susceptibility of TGG of 2×10^{-3} emu cm^{-3} Oe^{-1} (molar susceptibility of 0.1 emu mol^{-1} Oe^{-1}),[19] we calculate the corresponding effective magnetic field as $B_{\mathrm{eff}} = 0.5$ G. Then, applying Eq. (1) and taking l as the crystal thickness (as a maximum estimate[15]), one predicts a Faraday rotation of 2.3×10^{-2} mdeg. This prediction is in striking contrast with the measured value of 150 mdeg, i.e., a discrepancy of 4 orders of magnitude. This result completely invalidates the common assumption that the direct and inverse Faraday effects are determined by the same Verdet constant and suggests that Eq. (5) fails on ultrashort time scales.

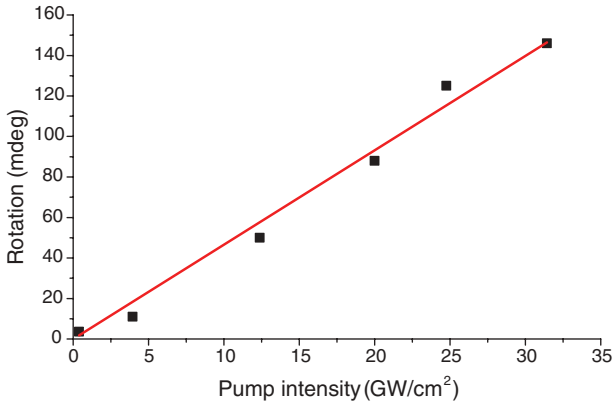

FIG. 4. (Color online) The Faraday rotation associated with the IFE contribution to the response is shown as a function of the pump peak intensity together with a linear fit.

V. DISCUSSION

The observed discrepancy between the measured magneto-optical response and the associated theoretical predictions is, by no means, trivial. Indeed, in the similar case of the electro-optical rectification, the nonlinear coefficient extracted from the femtosecond experiments agrees well with the values calculated from static measurements involving the Pockels effect.[20] Thus, the observed violation of the reciprocity between direct and inverse Faraday effects is specific to femtomagnetism.

Let us discuss possible origins of the observed discrepancy. One could argue that the postulated equality of the strengths of the direct and inverse Faraday effects is valid only in a medium without absorption.[7,9,10] However, the optical absorption bands of TGG are situated far from the experimental wavelength of 800 nm.[21] So, the absorption at 800 nm is $\sim 0.01 \ll 1$ and cannot account for the difference of 4 orders of magnitude observed in our measurements.

We speculate that the key reason for the observed discrepancy is related to the very short time scales of our experiments. This is corroborated by comparison with the results of Ref. 22 where the authors report optically induced magnetization in TGG at room temperature by measuring the voltage induced in a conducting coil attached to the crystal. The induced magnetic-flux density was determined as $\sim 10^{-4}$ G. The value is consistent with the predictions of Eq. (4) for the nonlinear magnetization induced in our experiments, noting that the peak intensity in Ref. 22 lies within the range of our lowest peak intensities. The measurements in Ref. 22 were performed using optical pulses with durations of several nanoseconds. The same authors also tried to measure the IFE in TGG in an all-optical pump-probe experiment with continuous-wave (cw) optical beams.[23] However, they concluded that the light-induced magnetization was too weak in the experiments to produce a detectable response. Our theoretical estimates presented above also predict that such a weak pump-induced magnetization would result in a response well below the sensitivity of our experimental apparatus. Nonetheless, we do observe the magneto-optical response, which is, therefore, due to the femtosecond duration of our pump pulses. Thus, we conclude that the phenomenological theory of Pershan is limited to the long time scales while failing in the subpicosecond regime.

Let us consider possible origins of the temporal dependence of the response. In the absence of a bias magnetic field, the magnetic dipoles of terbium ions (which are responsible for the paramagnetism of TGG) are disordered, and so the crystal does not have a net magnetic moment. The characteristic spin precession times associated with the paramagnetic ions are much longer than the time scales in the signal observed in our measurements. Thus, paramagnetic alignment is unlikely to occur during the action of the ultrashort laser pulse.[14,24] Hence, we postulate that the paramagnetism of TGG is unable to contribute significantly to the nonlinear magneto-optical response on femtosecond time scales. Yet, the tabulated Verdet constants are deduced from measurements of Faraday rotation caused by the alignment of paramagnetic ions due to an applied static magnetic field. Accordingly, the nonlinear magneto-optical susceptibility introduced in Eq. (2) has nothing in common with the (mainly paramagnetic) static Verdet constant of TGG.

A similar argument applies to Eq. (1): The tabulated value of the Verdet constant, which refers to the measurements of Faraday rotation caused by static alignment of paramagnetic ions, can hardly be used in the description of femtosecond phenomena.

Instead, we argue that the ultrashort optical pulse excites transitions to virtual states with high values of orbital momentum. The transient magnetization associated with the nonequilibrium angular momentum leads to the observed transient Faraday signal. Since this magnetization originates from the orbital motion rather than from the electronic spin, we call it "ultrafast diamagnetic magnetization."

As an aside, we note that the signal measured in the pump-probe experiment is a convolution of the direct and inverse Faraday effects and is, therefore, proportional to the Verdet constant squared. Hence, the experiment does not allow us to isolate the sign of the Verdet constant and thereby to differentiate between the diamagnetic and the paramagnetic contributions or to estimate their relative strengths.

Let us consider more closely the notion of a light-induced effective magnetic field, e.g., as introduced in Ref. 10 for cw light and more recently in Ref. 25 as a magnetic field induced by circularly polarized optical pulses,

$$H_{\text{eff}} = M^{\text{NL}}/\chi. \tag{6}$$

The field has been used in several recent papers.[26–33] Yet, it was demonstrated that the effective field model fails to predict the correct frequency of the light-induced magnetization precession in paramagnetic $Dy_3Al_5O_{12}$.[34] Furthermore, the most recent analyses show that the helicity-dependent magnetization switching in rare-earth transition metal alloys is not actually related to the effective field.[35] Moreover, one might be tempted to treat the effective field as a genuine Oersted field and, therefore, to substitute it into Maxwell equations or to use it in Eq. (1) as we did above. This not only leads to the quantitative inconsistency discussed above, but also cannot be justified in terms of Maxwell equations. Hence, the notion of the light-induced effective field should be used carefully. In particular, this field, which commonly refers to the torque exerted on spins via spin-orbit interaction enhanced due to the optically unquenched orbital momentum (responsible for the transient magnetization), does not produce an electric field via the Faraday induction law and does not act on electrical charges.

The effective magnetic field defined via Eq. (6) depends not only on the Verdet constant, but also on the susceptibility χ. However, the transient nonlinear magnetization has a spectrum that spreads from zero to a frequency of several terahertz. Hence, it is not obvious what susceptibility, which could also be modified via the intense optical excitation, should be used in Eq. (6). Also, the magnetic susceptibility loses physical meaning at such high frequencies.[36] The use of the light-induced effective magnetic field can be appropriate to describe the initial perturbation of the magnetization in magnetically ordered materials. However, it cannot align (almost instantaneously) paramagnetic moments in a disordered magnetic material, such as TGG. Neither should the light-induced effective magnetic field be associated with the optical pulse itself, e.g., such as in the notorious hypothesis of an axial dc magnetic field of a photon.[37,38] Indeed, the experimental observations from Refs. 22 and 23 disprove the latter idea completely.

The analogous case of the inverse electro-optical effect can be explained without introducing any effective electric field.[39] Indeed, there is no electric analog of the paramagnetic dipole moments. Thus, the electro-optical response is completely dielectric and is explained in terms of light-induced instantaneous nonlinear polarization. Since paramagnetic ordering cannot play a significant role in the direct and inverse Faraday effects in femtosecond pump-probe experiments, we conclude that the effects must be diamagnetic in nature, i.e., they do not involve "slow" spins. Moreover, as argued above, the strength of the effects on subpicosecond time scales might well be unrelated to the diamagnetic term in the static Verdet constant. The measured transient magnetization is created due to the rotating optical field and is related to the light-induced nonequilibrium orbital angular momentum. A theory of the IFE with particular attention to time scales was recently developed in Refs. 40 and 41, although the results did not directly apply to our measurements. Furthermore, when our paper was under review, our attention was drawn to Ref. 42, which was, however, unavailable in English. Judging from the abstract, equations, and figures in the article, it appears to pursue a goal of extracting third-order nonlinear susceptibility tensor components and is not directly related to our paper.

VI. SUMMARY

We present quantitative measurements of the ultrafast magneto-optical response of TGG at room temperature. The measured signal demonstrates behavior that is commonly attributed to the IFE. However, the amplitude of the measured Faraday rotation is at least 4 orders of magnitude greater than that predicted by the standard theory. Hence, our experiments support the idea that the phenomenological models of magneto-optical phenomena cannot be applied in the subpicosecond regime.

We argue that the ultrafast magneto-optical signal is determined by the instantaneous diamagnetic response rather than the paramagnetic alignment of spins by a light-induced effective magnetic field. Microscopically, the transient magnetization results from unquenching of the orbital momentum by the optically excited virtual transitions. The observed violation of the reciprocity between magneto-optical and optomagnetic phenomena results from the very different time scales associated with diamagnetic and paramagnetic mechanisms. Further studies, both theoretical and experimental, are required to clarify the nature of the magneto-optical response of paramagnetic dielectrics and, in particular, to resolve the relation between the inverse and the direct Faraday effects, something that cannot be achieved as a result of the presented measurements.

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