Optical magnetization, Part I: Experiments on radiant optical magnetization in solids

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Abstract: Linearly-polarized magnetic dipole (MD) scattering as intense as Rayleigh scattering is reported in transparent garnet crystals and fused quartz through a magneto-electric interaction at the molecular level. Radiation patterns in quartz show the strongest optical magnetization relative to electric polarization ever reported. As shown in an accompanying paper, quantitative agreement is achieved with a strong-field, fully-quantized theory of magneto-electric (M-E) interactions in molecular media. The conclusion is reached that magnetic torque enables 2-photon resonance in an *EH** process that excites molecular librations and accounts for the observed upper limit on magnetization. Second-order M-E dynamics can also account for unpolarized scattering from high-frequency librations previously ascribed to first-order collision-induced or third-order, all-electric processes.

References and links

1. C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (Dover, 1975).
2. A. Einstein and W. J. de Haas, "Experimental proof of the existence of Ampère's molecular currents," K. Akad. van Wet. Amsterdam. Proc. **18**, 696–711 (1915).
3. N. I. Zheludev and Y. S. Kivshar, "From metamaterials to metadevices," Nat. Mater. **11**(11), 917–924 (2012).
4. J. C. Ginn, I. Brener, D. W. Peters, J. R. Wendt, J. O. Stevens, P. F. Hines, L. I. Basilio, L. K. Warne, J. F. Ihlefeld, P. G. Clem, and M. B. Sinclair, "Realizing optical magnetism from dielectric metamaterials," Phys. Rev. Lett. **108**(9), 097402 (2012).
5. K. Y. Bliokh, Y. S. Kivshar, and F. Nori, "Magnetoelectric effects in local light-matter interactions," Phys. Rev. Lett. **113**(3), 033601 (2014).
6. N. A. Spaldin and M. Fiebig, "Materials Science. The Renaissance of Magnetoelectric Multiferroics," Science **309**(5733), 391–392 (2005).
7. P. Curie, "Sur la symétrie dans les phénomènes physiques, symétrie d'un champ électrique et d'un champ magnétique," J. Phys. **3**, 393–415 (1894).
8. P. Debye, "Remark on some new experiments on a magneto-electric straightening effect," J. Phys. **36**, 300–301 (1926).
9. H. B. Levine and G. Birnbaum, "Classical theory of collision-induced absorption in rare-gas mixtures," Phys. Rev. **160**(1), 159–169 (1967).
10. J. P. McTague and G. Birnbaum, "Collision-Induced Light Scattering in Gaseous Ar and Kr," Phys. Rev. Lett. **21**(10), 661–664 (1968).
11. J. A. Bucaro and T. A. Litovitz, "Molecular Motions in CCl4: Light Scattering and Infrared Absorption," J. Chem. Phys. **55**(7), 3585 (1971).
12. G. C. Tabisz, W. R. Wall, and D. P. Shelton, "Collision-Induced Light Scattering from Liquid CCl4 and C6H12," Chem. Phys. Lett. **15**(3), 387–391 (1972).
13. M. Perrot, M. H. Brooker, and J. Lascombe, "Raman light scattering studies of the depolarized Rayleigh wing of liquids and solutions," J. Chem. Phys. **74**(5), 2787 (1981).
14. See for example Y. R. Shen, See for example Y. R. Shen, *Principles Of Nonlinear Optics*, 1st ed. (John Wiliey & Sons, 1984), pp. 195–199.
15. W. K. H. Panofsky and M. Phillips, *Classical Electricity and Magnetism*, 2nd ed. (Addison-Wesley Publishing Co., 1962).
16. L. D. Landau, E. M. Lifshitz, and L. P. Pitaewskii, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon Press, 1984), pp. 268.
17. P. Pershan, "Nonlinear optical properties of solids: energy considerations," Phys. Rev. **130**(3), 919–929 (1963).

18. A. A. Fisher, E. F. Cloos, W. M. Fisher, and S. C. Rand, "Dynamic symmetry-breaking in a simple quantum model of magneto-electric rectification, optical magnetization, and harmonic generation," Opt. Express **22**(3), 2910–2924 (2014).
19. V. B. Berestetskii, E. M. Lifshitz, and L. P. Pitaevskii, *Quantum Electrodynamics*, 2nd ed. (Butterworth-Heinemann, 1982), Vol. 4.
20. A. A. Fisher, E. F. C. Dreyer, A. Chakrabarty, and S. C. Rand, "Optical magnetization, Part II: Theory of induced optical magnetization," Opt. Express **24**(23), 26076–26091 (2016).
21. S. L. Oliveira and S. C. Rand, "Intense Nonlinear Magnetic Dipole Radiation At Optical Frequencies: Molecular Scattering in a Dielectric Liquid," Phys. Rev. Lett. **98**(9), 093901 (2007).
22. S. C. Rand, W. M. Fisher, and S. L. Oliveira, "Optically induced magnetization in homogeneous, undoped dielectric media," J. Opt. Soc. Am. B **25**(7), 1106 (2008).
23. M. Fiebig, D. Fröhlich, T. Lottermoser, V. V. Pavlov, R. V. Pisarev, and H.-J. Weber, "Second harmonic Generation in the Centrosymmetric Antiferromagnet NiO," Phys. Rev. Lett. **87**(13), 137202 (2001).
24. C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and T. Rasing, "All-optical magnetic recording with circularly polarized light," Phys. Rev. Lett. **99**(4), 047601 (2007).
25. W. M. Fisher and S. C. Rand, "Optically-induced charge separation and terahertz emission in unbiased dielectrics," J. Appl. Phys. **109**(6), 064903 (2011).
26. S. C. Rand, "Quantum theory of coherent transverse optical magnetism," J. Opt. Soc. Am. B **26**(12), B120 (2009).
27. N. Minkovski, G. I. Petrov, S. M. Saltiel, O. Albert, and J. Etchepare, "Nonlinear polarization rotation and orthogonal polarization generation experienced in a single-beam configuration," J. Opt. Soc. Am. B **21**(9), 1659 (2004).
28. R. Krsmanović, V. A. Morozov, O. I. Lebedev, S. Polizzi, A. Speghini, M. Bettinelli, and G. V. Tendeloo, "Structural and luminescence investigation on gadolinium gallium garnet nanocrystalline powders prepared by solution combustion synthesis," Nanotechnology **18**(32), 325604 (2007).
29. C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (J. Wiley & Sons Inc., 1992).

1. Introduction

In molecular liquids and solids, it is well-known that an exchange of angular momentum can take place between orbital (or spin) degrees of freedom and rotations (or librations) due to the fact that they are coupled [1]. An early demonstration of this was provided by the Einstein-de Haas experiment [2]. Yet, it has never been suggested that the magnetic field of light could directly drive such an exchange between internal and external momenta efficiently through a simple torque interaction at the molecular level. Neither has any dynamic interaction been shown to produce magnetization rivalling the electric polarization that can be excited at frequencies near allowed atomic transitions in homogeneous media. However in this paper we report experiments in which the intensity of induced magnetic dipole scattering equals that of electric dipole scattering ($I_{ED} = I_{MD}$), and a 2-photon magneto-electric interaction is analyzed that accounts for it. We show that by imparting angular momentum to a system with an initial photon, light can subsequently convert orbital motion to rotational motion by the action of magnetic torque from a second photon. This makes enhanced optical magnetism possible in homogeneous dielectrics without requiring any exotic structure or rare material properties. Thus it offers an alternative to specialized media such as metamaterials [3,4] or magneto-electric multiferroics [5,6] for the realization of high frequency magnetism. Furthermore, the mechanism of induced magnetization that accounts for the present results has the unique feature of being proportional to the electric polarization and does not rely on intermediate fields or couplings such as one finds in ferromagnetic, ferroelectric, ferroelastic, or piezoelectric materials. Consequently, to our knowledge, this is the first observation and explanation of a magnetoelectric interaction [5,7,8] at the molecular level.

The experimental results presented here report light scattering intensities recorded in a ninety-degree geometry for various input polarizations and intensity levels. A considerable literature exists on depolarized spontaneous light scattering in gases and condensed matter at low intensities. Cross-polarized scattering in the wing of the Rayleigh line was first reported in rare gases [9,10] and was ascribed to anisotropies induced by collisions that led to linear light scattering. Collisions were theorized to distort molecular charge distributions sufficiently that unpolarized ("depolarized") electric dipole scattering of light was induced.

Indeed a quadratic dependence on gas density was reported, pointing to the importance of binary collisions in the experiments. In condensed matter systems, such as liquid CCl_4 composed of spherical top molecules, depolarized Rayleigh scattering was similarly observed and interpreted as being of collisional origin, although disparities were noted in the light scattering spectrum between fitting parameters for low and high frequencies [11,12]. Later authors emphasized that at least in anisotropic molecular systems the induced charge motions involved external coordinates of the molecules composing the medium rather than internal coordinates. That is they were librational (rotational) rather than vibrational in character [13]. This was certainly true at high pump intensities in anisotropic systems like benzene where electric torque could be exerted on induced polarization anisotropy via third-order (nonlinear) light scattering to cause rotational excitation [14]. Significantly however, the depolarization ratio was assumed to be constant at low to intermediate intensities and detailed measurements of polarization patterns of scattered light were never undertaken. The presence of an intensity-dependent dipolar component polarized at right angles with respect to the incident polarization was completely overlooked, as neither radiation patterns nor intensity dependences were recorded in these early experiments. Librational excitations were assumed to originate from random "collisions" with a fixed depolarization ratio or from all-electric interactions of third- or higher-order in isotropic and anisotropic molecular systems alike. In this paper, detailed measurements of radiation patterns and intensity dependence of quasi-elastic light scattering, together with new theoretical considerations, have permitted us to draw very different conclusions regarding the mechanism of cross-polarized scattering at moderate intensities in dense molecular liquids and simple dielectric solids.

On the basis of the well-known multipole expansion [15], the proportion of light scattered from induced magnetic dipoles (MD) is expected to be negligible compared to that from electric dipoles (ED) in natural materials ($I_{MD} / I_{ED} << \alpha^2$, where α is the fine structure constant). In the past, it was even argued that electric polarization should vanish merely to enable the detection of weak magnetic effects at optical frequencies [16]. Still, detectable magnetic fields are known to result from the inverse Faraday effect, in which circularly-polarized light generates static magnetization along the direction of propagation. Magnetic response can also be elicited from carefully-engineered inhomogeneous media at high frequencies [3,4]. However, in such cases it is the electric not the magnetic field that drives the response. For this reason it is surprising to find, as reported here, that a nonlinear magnetization process driven jointly by optical electric and magnetic fields according to $M = \chi EH^*$ not only takes place in homogeneous, transparent optical materials at the molecular level, even when conventional symmetry analysis forbids it [17], but can be remarkably intense.

Radiant magnetization is an allowed process in homogeneous media, due to the fact that parity is not conserved and time-reversal invariance is not obeyed in magneto-electric interactions. M-E interactions are quite unusual in this regard and are subject to combined parity-time (P-T) symmetry [5,18], a special case of the more general CPT symmetry governing physical processes [19] when charge is conserved. The unexpectedly large magnitude of optically-induced magnetization reported here at moderate intensities may be accounted for by an analysis (see companion paper II, Ref [20].) in which magnetic torque alters the areas enclosed by charge motion in optically-excited molecular systems. The earliest evidence of a molecular-scale process of this kind was reported in transparent molecular liquids [21,22] but here magnetization mediated by the optical Lorentz force is reported that is as intense as first order electric polarization in solids. The experimental results are well-explained with the exactly solvable model developed in II. We show that optical magnetic torque not only mediates an unforeseen type of electronic transition but increases the area enclosed by librational electron motion within individual molecules, thereby enhancing the magnetic moment driven jointly by the E and H fields of light. A universal

mechanism is therefore identified for enhancing high frequency magnetic phenomena which should catalyze advances on established research topics such as magnetic domain imagery [23], ultrafast magnetic data storage using ultrafast pulses [24] and negative permeability in dielectric materials [4]. These findings may also lead to novel methods of converting electromagnetic energy directly to electricity with negligible heat production, generating THz radiation in unbiased insulators, or producing large (oscillatory) magnetic fields without current-carrying coils [25,26].

2. Experimental results

Our experimental procedures have been described previously [22] but are summarized here for convenience. Pulsed laser systems of two types were used to record light scattering signals together with their radiation patterns (polar power plots) obtained by rotating the incident polarization in the 90 degrees geometry of Fig. 1. One laser was a Clark-MXR CPA-2001 that delivered amplified ~150 fs pulses of wavelength 775 nm at a rate of 1 kHz and the other was a cw mode-locked oscillator (Coherent Vitara) emitting ~20 fs pulses at a wavelength of ~800 nm and a repetition rate of 80 MHz. Automatic dispersion compensation was provided with a wavefront controller (Biophotonic Solutions MIIPS) to ensure reproducible peak intensities in the interaction volume. The interaction volume was small and contained no surfaces. Scattered light intensity was recorded through an interference filter with a rectangular passband of ±2.5 nm centered on the laser wavelength using a photomultiplier or an FND-100 photodiode in the kHz and 80 MHz setups respectively. The filter excluded vibrational Raman scattering and impurity fluorescence from the measurements. The input laser polarization was rotated under computer control to map out radiation patterns of the scattered light for fixed analyzer orientations, and an adjustable attenuator permitted studies versus input intensity. Typically an acquisition time of 20 minutes was required to record data for two orientations of the analyzer at each input intensity. Scans were performed over two decades of intensity, centered on a value determined by magnification of the collimated beam and limited by the range of the input attenuator.
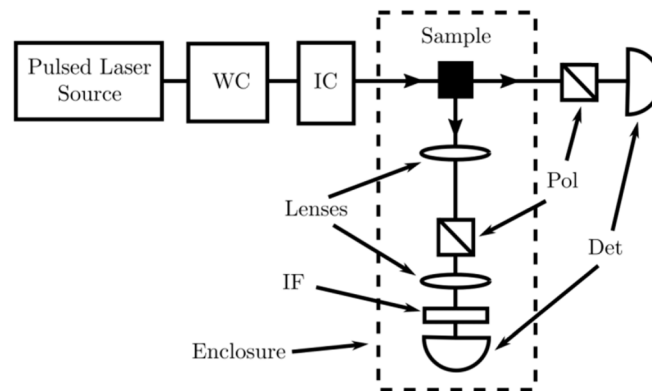

Fig. 1. Schematic diagram of the experimental apparatus, showing both the forward beam used to monitor polarization changes due to cross-polarized four-wave mixing in sample transmission and the detection arm at 90 degrees to the incident beam used to observed ED and MD scattering. WC = wavefront controller, IC = Intensity Controller, IF = interference filter, Det = detector, Pol = polarization analyzer.

All measured radiation patterns were decomposed into polarized and unpolarized components. This resulted in four distinct data sets for each material studied, yielding vertical and horizontal dipolar components (denoted ED and MD respectively) and their corresponding unpolarized background signals. Quadrupolar moments should be smaller than ED moments by 5.5×10^{-6} or contribute to scattering at a level below 3×10^{-11} of the

intensities measured here for small molecules and consequently can be ignored. The result of this decomposition is shown in Fig. 2(a) for liquid CCl_4, which served as a reference sample. In CCl_4, fitted curves showed that all signal components were quadratic with respect to input intensity except for the polarized Rayleigh signal, which was linear as expected. The polarized components in CCl_4, which is a non-chiral and optically isotropic liquid at room temperature, were purely dipolar and orthogonal to the incident polarization (see also [21,22]).
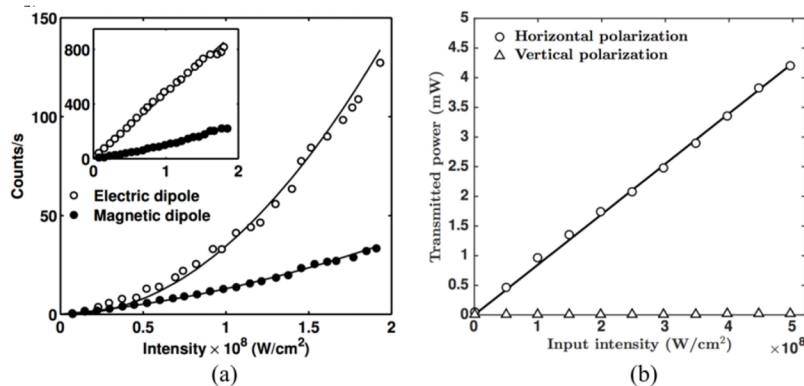

Fig. 2. (a). Measurements of unpolarized ED (open circles) and MD (filled circles) scattering intensity versus input intensity for the reference sample (CCl_4) obtained at a repetition rate of 1 kHz. Solid curves are quadratic fits to the intensity dependence. Inset: corresponding data for polarized scattering components on the same intensity scale. (b) Measurements of sample transmission in GGG with polarization parallel (circles) and perpendicular (triangles) to that of the incident beam in the range of input intensities used for scattering experiments.

Additionally, polarization analysis of the forward beam transmitted by the sample showed that no ellipse rotation or cross-polarized four-wave mixing [27] took place in the intensity range of our experiments (Fig. 2(b)). Hence the scattering in CCl_4 that was polarized perpendicular to Rayleigh scattering can only have arisen from second-order magnetization at the molecular level. Because all-electric, second-order nonlinearities are prohibited in centrosymmetric compounds, whereas processes driven jointly by E and H are not [18], it may further be concluded that the mechanism behind this phenomenon is magneto-electric. The quadratic intensity dependence of the unpolarized electric and magnetic "background" signals in Fig. 2(a) indicate that they too were the product of a second-order, magneto-electric interaction.

Next, radiation patterns were recorded for Gadolinium Gallium Garnet (GGG). GGG is a body-centered cubic crystal with space group $O_h^{10} = Ia\bar{3}d$ that includes inversion [28]. Furthermore, the sample contains no internal domain structure or magnetization and any symmetry-breaking that occurs is a dynamic rather than a static process. Hence, like the reference sample, it does not support second-order, all-electric nonlinearities. Yet, as indicated by the raw data of Fig. 3(a), a strong cross-polarized radiation pattern is observed in the range $10^7 - 10^9$ W/cm^2. Figure 3(b) shows that the same components of the GGG data exhibited a quadratic intensity dependence as in the reference sample CCl_4. The polarized signal component orthogonal to the Rayleigh radiation pattern can therefore be attributed to magnetic dipole (MD) scattering. However its magnitude in this intensity range was somewhat less than that of the Rayleigh component. The ratio of unpolarized MD and ED scattering intensities was similarly less than unity $(I_{MD} / I_{ED} \sim 0.9)$. At a higher intensity $(2 \times 10^{10}$ W/cm$^2)$ in both GGG and fused quartz however the ratio of polarized intensities reached an upper limit of unity $(I_{MD} / I_{ED} = 1)$ (see Fig. 4). To our knowledge, this induced

MD effect is the most intense with respect to linear ED polarization ever observed, and it greatly exceeds expectations from the multipole expansion.
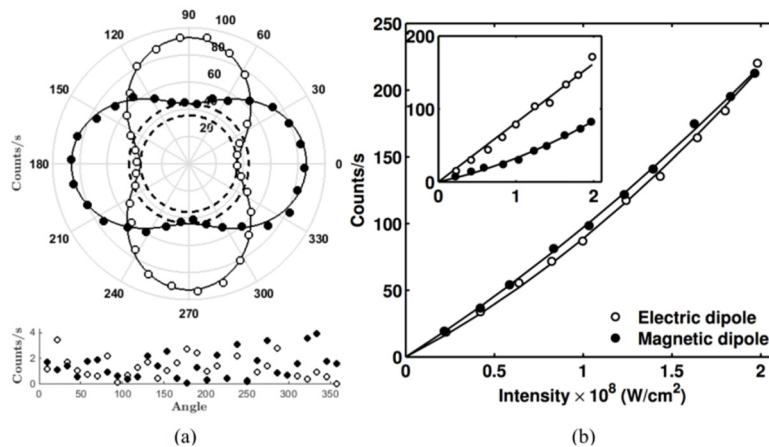

Fig. 3. (a) Polar plot of raw data on co-polarized (open circles) and cross-polarized (filled circles) radiation patterns in GGG at $I = 1.4 \times 10^7$ W/cm^2 obtained at a repetition rate of 1 kHz. Dashed circles anticipate fits to the unpolarized background signal intensities. Residuals from the best fit of a circle plus a squared cosine curve to the raw data are shown below the polar plot. (b) Comparative plots in crystalline GGG of unpolarized ED (open circles) and MD (filled circles) scattering. Solid curves are quadratic fits to the data. Inset: corresponding data for polarized scattering components on the same scale.
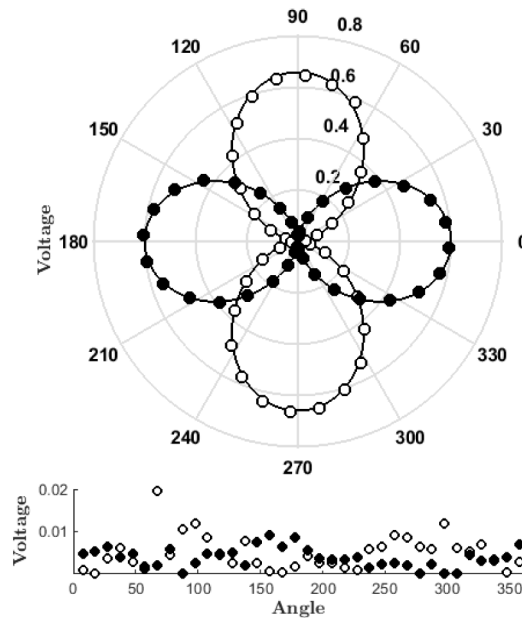

Fig. 4. Polar plots of the radiation patterns for polarized ED and MD scattering in fused quartz at an input intensity of $I \sim 2.2 \times 10^{10}$ W/cm^2 obtained at a repetition rate of 80 MHz. At this intensity, in this sample, the unpolarized component is negligible compared to the polarized component. Note that peak intensities in the two plots are equal. Residuals from the best fit of a squared cosine curve to the raw data are shown below the polar plot.

These experimental results may be analyzed by considering a nominally 2-level molecular system subjected to strong electric and magnetic driving forces. In companion paper II, a

dressed state approach [29] is developed that extends prior analysis of weak (but non-vanishing) magneto-electric interactions in atoms [18] to molecules. The analysis of Ref. 18 revealed that dynamic symmetry-breaking makes possible the radiant magnetization probed by our scattering experiments, but the induced moments were predicted to be extremely weak. Only in II are the magnitudes of optically-induced electric and magnetic dipoles equal in the high intensity limit. In a key departure from the atomic model, internal and external degrees of freedom of charge motion in a homonuclear diatomic molecule are explicitly taken into account in II. For simplicity spin is ignored, but orbital motion and molecular rotation are included in the total angular momentum ($J = L + O$ [1]), and it is pointed out that an exchange of angular momentum between them can be affected by magnetic torque. The introduction of magnetic torque in the excited state of the molecule is a vital aspect of the interaction, for it mediates the transfer of orbital to rotational angular momentum, admixes angular momentum into the ground state and allows the magnetic field to de-excite the molecule. It thereby enables a nearly resonant magnetic transition at the optical frequency. At the same time, the effective area enclosed by angular motion (libration) of the electron increases, accounting for enhancement of the magnetic moment at the molecular level.

3. Discussion and conclusions

The main conclusions in II are substantially confirmed by the present experimental results. In the low power limit, the theoretical dependence of radiant magnetization on input intensity is predicted to be quadratic. This agrees with the experimental results in Figs. 2(a) and 3(b). The frequency of magnetic scattering in Eq. (4) of II is predicted to be the optical frequency. This too agrees with experiment, since our narrow detection passband is centered on the laser wavelength. However the width of the passband is sufficient to allow transmission of scattered light that is shifted by librational frequencies of our samples. Consistent with this, the presence of two (polarized and unpolarized) components in each radiation pattern of our data provides indirect evidence for the presence of quasi-elastic components in the scattered light. In Fig. 5, two magnetic transitions are shown to be possible during the magneto-electric interaction when sufficient bandwidth is available in the laser source. The solid downward arrow in the figure depicts a polarization-preserving transition at the center frequency ω of the incident light. The dashed arrow depicts a second transition at a shifted frequency $(\omega - \omega_\varphi)$ which terminates by resonantly exciting a rotation of frequency ω_φ. By exciting molecular rotation this process should cause unpolarized scattering, consistent with our observations of two components in each radiation pattern (seen most clearly in the raw data of Fig. 3 where the relative intensities of unpolarized and linearly-polarized components are comparable). Finally, in the high intensity limit, as shown by the calculations of paper II, magneto-electric interactions can induce MD moments as large as ED moments. This agrees quantitatively with the fully saturated response observed in quartz at elevated intensities (Fig. 4).

In the past, stimulated Rayleigh-wing scattering (a third-order interaction involving electric torque) has been invoked as the cause of unpolarized, rotationally-averaged, scattering in anisotropic molecular liquids [14]. However electric torque cannot develop in CCl_4, GGG or fused quartz due to their isotropy. Differential polarizability is precluded in spherical top molecules and the isotropic solids studied here. Collisionally-induced anisotropy similarly cannot explain our results. Collisional effects produce a fixed depolarization ratio, unlike the data of Fig. 2 that reveals a quadratic intensity dependence in CCl_4. Additionally, neither spontaneous nor stimulated electric-field-induced scattering can account for the strong polarized component evident in quartz at high intensity (Fig. 4, filled circles). Thus the second-order scattering reported here and its decomposition into polarized and unpolarized components (Figs. 2(a) and 3(b)) must have a different origin, which we ascribe to the two downward magnetic transitions in Fig. 5, as discussed above. This points to a new mechanism for the generation of rotations in molecular systems, and of librations in condensed matter,

provided the imputed torque interaction analyzed in II is ultrafast under the conditions of our experiments. To determine the speed with which optical torque can act, we estimated the time interval τ necessary for the projection m_ℓ of angular momentum on the quantization axis to change by one unit due to magnetic torque at the intensities used in our experiments. By solving the quantum mechanical torque equation (see II), the time interval was found to be $\tau = \hbar / \mu_0^{(e)} E \approx 125$ fs for optical fields of $E \approx 10^8$ V/m. Hence for the field strengths and pulse durations of the present experiments, the dynamic torque interaction theoretically goes to completion during each pulse.
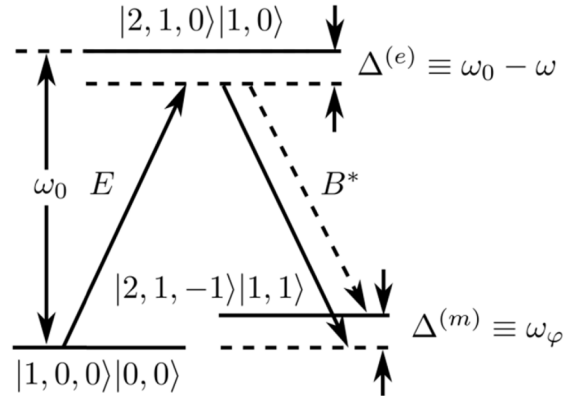

Fig. 5. An energy level diagram depicting the second-order magneto-electric dynamics driven by E and B fields of (i) equal frequency (solid arrows), and (ii) frequencies differing by rotational frequency ω_φ (dashed arrow).

As detailed in II, the unexpected and comparatively large magnetic scattering reported here can be accounted for by magnetic torque dynamics in the excited states of molecules at the field strengths used in our experiments. The essential perspective provided in paper II is that at intermediate fields magnetic torque on orbital momentum causes conversion of orbital to rotational angular momentum embodied in librations about the center-of-mass rather than about the molecular axis. The area associated with librational (rotational) motion is much larger than that of the initial excited state orbital. Hence the molecular magnetic moment m=IA , which is proportional to current and area, as well as the scattering intensity are enhanced to the point of equaling electric dipole effects. The main result of the present work is the experimental finding that cross-polarized nonlinear scattering, interpreted here to be induced magnetic dipole scattering, can indeed equal electric dipole scattering in intensity (Fig. 4). Optically-induced magnetization of this magnitude in unprecedented and, as far as we know, has neither been observed nor predicted in past work. The fact that cross-polarized scattering has not previously been analyzed as nonlinear magnetization in light-scattering studies of the Rayleigh wing may be attributed to the absence of measured radiation patterns, measured intensity dependences, or the use of high intensity light in earlier work.

Our interpretation of the experimental results ignores possible contributions from electric quadrupole moments. This may be justified by the standard multipole expansion in which quadrupolar contributions are smaller than electrical dipole terms by a factor of the square of the wavenumber times the particle radius, estimated to be $(ka)^2 < 5.5 \times 10^{-6}$ here. This is inconsistent with the data of Fig. 4 where the ratio of MD and ED polarized signals is one. Also, surface contributions have been ignored because there were no interfaces in or near the scattering volume in our experiments. Finally, our results are not due to internal symmetry-breaking from magnetic domain structure. All samples were free of internal magnetic order

and it is particularly noteworthy that our reference liquid CCl$_4$ and fused quartz sample are both "non-magnetic" in the conventional sense.

In summary, the upper limit of magnetic dipole scattering intensity in solids predicted in II has been confirmed to equal that of Rayleigh scattering at moderate intensities. This implies that MD moments induced by light can be as large as ED moments as the result of magneto-electric dynamics originating from a saturated second-order nonlinear optical process at the molecular level. Observations of optical magnetization in this work agree in polarization, frequency, intensity-dependence, and saturation behavior with a model of magneto-electric interactions terminating in the ground electronic state of molecules as the result of magnetic torque. Hence a "ground state" molecular model (that of paper II) appears to explain enhanced optical magnetism observed via quasi-elastic scattering in natural materials in the present work. Depolarized scattering also appears to be caused by the magneto-electric process when ultrafast pulses of sufficient bandwidth are used for excitation, pointing to a novel mechanism for generating oriented rotations. We anticipate that in atomic systems, it may be possible for magnetic torque to act on spin to produce enhanced magnetic effects in the same way that we conclude here that it has acted on orbital angular momentum.