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Dependence of optically induced magnetism on molecular electronic structure

W.M. Fisher *, S.C. Rand

Division of Applied Physics, University of Michigan, Ann Arbor, MI 48109-1022, United States

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ABSTRACT

Intense magnetic dipole scattering that has been observed recently in several transparent insulators arises nonlinearly and saturates as predicted by classical theory, but also varies with electronic structure of the medium.

1. Introduction

In transparent dielectrics, spontaneous optical scattering from magnetic dipole interactions is typically five orders of magnitude less intense than electric dipole scattering. However, ultrafast nonlinear dynamics can mediate strong magnetic response via a quadratic nonlinearity at intensities well below the relativistic range. At input intensities in the range 10^7–10^8 W/cm^2, we report magnetic dipole scattering that is one quarter as intense as electric dipole scattering and depends on molecular structure.

Some aspects of this optically induced magnetism are universal, such as the maximum magnetic dipole moment relative to the electric dipole moment. As a consequence, the highest ratio of magnetic to electric scattering intensity is found to be 0.25 in three transparent dielectric liquids. Other properties, such as the slope with which magnetic response rises with respect to electric response is predicted (and observed) to vary with electronic structure of the scattering medium. In this paper we demonstrate that the ratio of MD/ED scattering intensities, which is linear in all transparent dielectrics studied to date up to its saturation value, grows at different rates in samples of water, CCl_4, and benzene. This reflects differences in the intra-molecular potentials experienced by bound electrons, and implies that potential applications of this phenomenon to the modification of refractive indices, optical magnetic resonance, optical isolation and communication will respond to structural selection or optimization, although the media in question are homogeneous and "non-magnetic".

2. Experiments

We recorded polarized light scattering signals using a 90° scattering geometry in three insulators. The incident beam was the amplified, pulsed output from a Clark-MXR 2001 laser system, nominally consisting of 100 fs linearly polarized pulses at a repetition rate of 1 kHz. Measurements were performed using the same approach outlined in previous publications [1,2], except that the input power was varied using a precision-compensating wedge device to avoid beam deviation and nonlinearities from the intensity controller. Polarization of the input beam was controlled with a half-wave plate placed before the sample.

Scattered light intensities were observed through limiting apertures and a linear polarization analyzer which removed electric contributions of all orders. A low-noise photomultiplier was used for detection, with a lower limit on ratio measurements set by an effective contrast of 1.5% between orthogonal polarizations that accounted for small angular misalignments, finite polarizer contrast, and the degree of polarization of the input beam. The input power was then varied, with results that are summarized in Fig. 1 for experiments in CCl_4, benzene, and water. All measurements were made at room temperature in samples that were double filtered at the 0.1 μm level.

* Corresponding author. Tel.: +1 734 936 0181.
E-mail address: wmfisher@umich.edu (W.M. Fisher).

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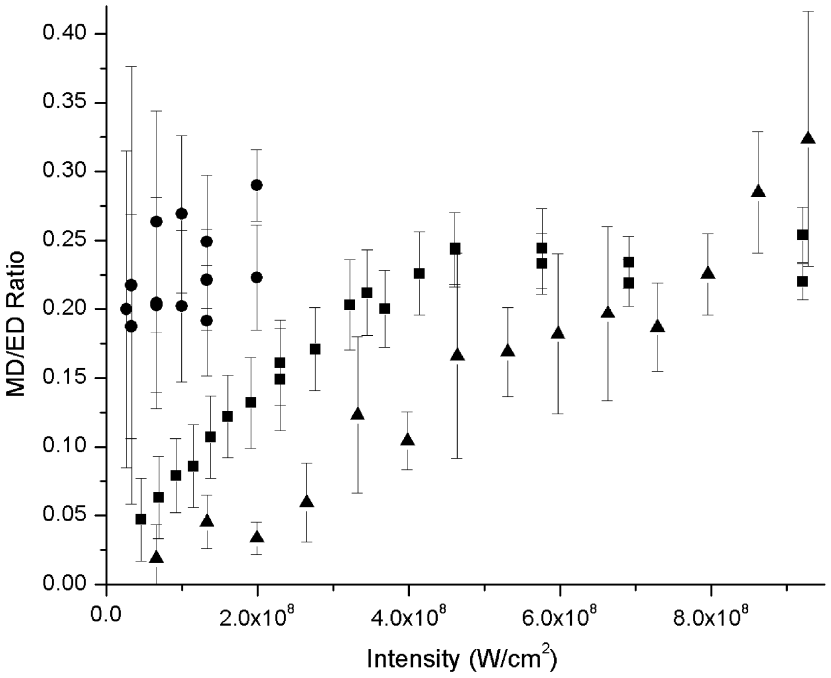

Fig. 1. Ratio of magnetic to electric dipole scattering intensities in water (triangles), CCl_4 (squares), and benzene (circles) versus incident laser intensity.

3. Analysis

The upper limit for radiant magnetic dipole intensity was calculated in an earlier paper, using an argument based on Ampere's law [1] and a continuum model for the charge distribution. There it was shown that up to 25% of scattered light can be magnetic in origin at non-relativistic intensities. To make additional predictions requires a more detailed model however.

The complex nonlinear dynamics initiated by weak magnetic Lorentz forces in bound electron systems can be modelled accurately using a Hooke's law model. Provided the optical frequency is not close to any electronic resonances, we may simply analyze Newtonian equations for the motion induced by a plane-wave polarized along $\hat{x}$ and propagating along $\hat{z}$ through a uniform distribution of bound charges, initially at rest. The external electric and magnetic field components are $\bar{E}(z,t) = (1/2)\hat{x}(E_0 \exp[-I(\omega t - kz)] + c.c.)$ and $\bar{B}(z,t) = (1/2)\hat{y}(B_0 \exp[-I(\omega t - kz)] + c.c.)$. Induced charge motions in the $\hat{x}$ and $\hat{z}$ directions obey the force equations

$$\frac{\partial^2 x(t)}{\partial t^2} + \gamma_1 \frac{\partial x(t)}{\partial t} + \omega_1^2 x(t) = -\frac{eE(t)}{m_e} + \frac{eB(t)}{m_e}\frac{\partial z(t)}{\partial t}, \tag{1}$$

and

$$\frac{\partial^2 z(t)}{\partial t^2} + \gamma_2 \frac{\partial z(t)}{\partial t} + \omega_2^2 z(t) = -\frac{eB(t)}{m_e}\frac{\partial x(t)}{\partial t}. \tag{2}$$

The force constants K_1 and K_2 correspond to orthogonal motions along $\hat{x}$ and $\hat{z}$, respectively, and are assumed to be unequal ($K_1 \neq K_2$). Similarly, the damping coefficients of these motions γ_1 and γ_2 are assumed to be unequal ($\gamma_1 \neq \gamma_2$).

By solving for the amplitudes of induced motion along x and z, and using them to write expressions for the corresponding electric and magnetic currents (J_E and J_M respectively), radiant electric and magnetic intensities can ultimately be compared. When this is done, one finds

$$R = |I_M/I_E| = \frac{4\omega^2\omega_c^2[(4\omega^2 - \omega_0^2)^2 + (2\omega\gamma_0)^2]}{[(4\omega^2 - \omega_0^2)^2 + 4\omega^2\gamma_0^2]^2}, \tag{3}$$

where ω is the optical frequency, $\omega_c \equiv eB/m_e$ is the cyclotron frequency, and ω_0 and γ_0 are the frequency and linewidth of the magnetic resonance, respectively. When conditions for parametric resonance are met (i.e. high input power and $4\omega^2 - \omega_0^2 \approx 0$), the intensity ratio of scattered magnetic to electric radiation varies as the square of the cyclotron frequency, since then $|I_M/I_E|_{res} = (\omega_c/\gamma_0)^2$. This ratio is linear in the incident intensity up to saturation, since it can be re-written as

$$\frac{I_{MD}}{I_{ED}} = \left[\left(\frac{e}{m_e}\right)^2 \frac{n\sqrt{\mu_0\mu}}{c_0\gamma_0^2}\right] I_{inc} \tag{4}$$

n is the refractive index and c_0 is the speed of light in vacuum. e/m_e is the charge-to-mass ratio of the electron and μ is the permeability of the medium. I_{inc} is the intensity of the pump wave.

4. Results

The results of Fig. 1 show excellent agreement with both the intensity dependence of non-resonant magnetic dipole scattering predicted in Section 3 and its limiting value. In water and CCl_4, the saturation level of $R = 0.25$ is achieved above intensities of roughly 5×10^8 W/cm^2, whereas this level of response is already achieved in benzene at much lower intensities. In addition , the intensity dependence (observed clearly in the data for water and CCl_4 below 5×10^8 W/cm^2) is linear in the input intensity, as predicted by Eq. (4) under conditions of parametric resonance. Although the optical frequency was not chosen to meet a specific tuning condition like $4\omega^2 - \omega_0^2 = 0$, and in fact ω_0 may not be equal to 2ω in the transparent liquid samples of our experiment, it is well known that essentially any frequency will excite primary parametric resonances when the modulation index is sufficiently high [3]. Hence, we assume the apparent frequency tuning requirement is theoretically relaxed by simply increasing the forcing field strength.

From Eq. (4) it can be seen that materials with a large index of refraction should have relatively large magnetic response if their magnetic resonance linewidths are comparable. However,

the squared reciprocal dependence on magnetic linewidth is expected to be the dominant factor in determining the slope of R versus I_{inc}. While our observations in water, CCl_4, and benzene by themselves make it clear that structural differences alter magnetic response at optical frequencies, quantitative comparisons between theory and experiments will have to await independent determinations of magnetic resonance frequencies ω_2 and linewidth factors γ_2 in different materials.

Magnetism at optical frequencies can be very intense through parametric enhancement. Our observations indicate that enhancement by five orders of magnitude takes place above $\sim 10^7\,W/cm^2$. Here we have shown that magnetic intensities rise quadratically as a function of input power in several dielectric liquids, saturate at a universal value of one quarter the intensity of electric dipole scattering, and are more intense in some materials than others at a given incident power, as the result of electronic structure variations. Measurements of magnetic linewidth factors are unavailable as yet, but the torsional damping parameter of molecular potentials is probably the most influential factor in determining the induced magnetic response.

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