Anomalous Anisotropic Light Scattering in Ge-Doped Silica Glass

P. G. Kazansky,[1,2,*] H. Inouye,[1] T. Mitsuyu,[1] K. Miura,[1] J. Qiu,[1] and K. Hirao[1,3]
[1]*Hirao Active Glass Project, ERATO, Keihanna Plaza, 1-7 Hikaridai, Seika-cho, Kyoto 619-02, Japan*
[2]*Optoelectronics Research Centre, University of Southampton, Southampton SO17 1BJ, United Kingdom*
[2]*Department of Material Chemistry, Graduate School of Engineering, Kyoto University, Sakyo-ku, Kyoto 606-01, Japan*

F. Starrost
Institute of Theoretical Physics and Astrophysics, University of Kiel, D-24098 Kiel, Germany
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Light scattering which peaks in the plane of light polarization is observed in glass pumped by intense laser radiation. The phenomenon is interpreted in terms of the angular distribution of photoelectrons in isotropic solid state materials. [S0031-9007(99)08624-X]

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Interaction of radiation with matter attracts considerable interest in many areas of science. In particular, since the discovery of the laser, many studies have been carried out on the interaction of intense laser radiation with glass, which is a key material in modern optical technology. Applications of glass span in the range from high power lasers for laser fusion [1] to optical waveguides for optical communication [2]. Light scattering, a phenomenon that occurs widely in nature, is also very common when intense laser beam propagates in optical glass materials [3]. It is well known that the scattering of polarized light in the plane of light polarization in an isotropic medium, such as glass, is always weaker compared to the orthogonal plane, since a dipole does not radiate in the direction of its axis. In this Letter, we report the observation of a new phenomenon in glass pumped by intense laser radiation—the scattering of light, in particular luminescence, which peaks in the plane of light polarization (anomalous anisotropic light scattering). The phenomenon is interpreted in terms of the angular distribution of photoelectrons in isotropic solid state materials.

In our experiments we used Ge-doped silica glass ($Ge:SiO_2$) which is the main constituent material of optical fibers for optical communication and which has an interesting property, strong photosensitivity, associated with defects in glass such as germanium oxygen deficient centers (Ge-ODC or Ge-Si wrong bonds) [4,5]. These centers produce a strong absorption band at 5 eV (240 nm, singlet-singlet transition), a weak absorption band at 3.7 eV (330 nm, forbidden singlet-triplet transition) and blue triplet luminescence at 3.1 eV with a decay time of about 100 μs [6,7]. Many studies of the blue luminescence in silica glass under the excitation with ultraviolet light have been carried out [8–10]. These studies have been motivated by the important role of defects responsible for the blue luminescence in two kinds of phenomena. The first one includes refractive index changes and gratings induced via one- or two-photon absorption of ultraviolet or visible light [5,11]. The second one involves gratings of second-order optical nonlinearity [12–14] induced via coherent photocurrent in glass [15] (modulation of the angular distribution of photoelectrons [16–18]) or coherent photoconductivity in glass under applied dc electric field [19] (modulation of the total cross section of ionization) as a result of quantum interference between coherent light fields at two different frequencies. More recently, strong refractive index charges were induced in glass by femtosecond laser pulses via multiphoton absorption of infrared light [20]. On the other hand, the luminescence of silica glass under excitation with intense (10^{11-12} W/cm^2) ultrashort (100 fs) infrared radiation has received little attention. It should be pointed out that high optical damage threshold of silica pumped with ultrashort pulses offers unique possibilities for the studies of optical excitations at high light intensities.

The laser radiation in Gaussian mode produced by regeneratively amplified mode-locked (120 fs pulse duration, 200 kHz repetition rate) Ti:sapphire laser operating at a wavelength of 800 nm was used in the experiments. Glass samples of $\sim$3 mm thickness were placed on a stage under the optical microscope (Fig. 1). The infrared laser radiation reflected by a dichroic mirror inside of the microscope was focused via a 20$\times$ objective onto the sample. The pump spot size in the focus of the beam was 4.6 μm. Simultaneously, the irradiated spot was imaged in the visible spectral range via the microscope by using a color CCD (charge-coupled device) camera.

During the experiments on Ge-doped ($GeO_2 \sim$ 8 mol%) silica glass strong blue luminescence (with a center wavelength at 410 nm) of defect states (Ge-ODC) was observed (Fig. 2). Using a cross-sectional area for Ge-ODC of 5×10^{-18} cm^2 [21], the absorption value at 240 nm for our sample of $\sim$15 cm^{-1} gives the Ge-ODC concentration of about 10^{19} cm^{-3}. This luminescence (triplet luminescence) can be excited via the singlet-singlet transition by absorption of three pump photons or one UV photon of the third harmonic of the pump

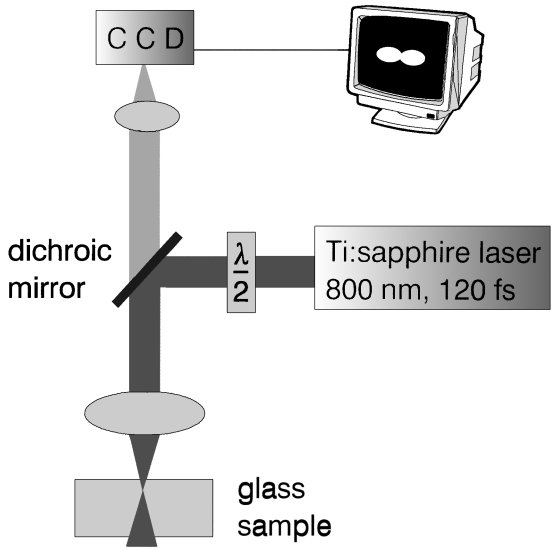

FIG. 1. Schematic of the experimental setup.

followed by quick nonradiative decay (with a decay time 1 ns) to the long-lived triplet level (Fig. 3).

When the pump (10 mW average power, 0.4 MW peak power, 2.5×10^{12} W/cm^2 intensity in the focus of a beam) was focused slightly ($\sim$50 μm) above the surface of the sample the shape of the spot of the blue luminescence imaged via the microscope and CCD camera was circular [Fig. 4(a)]. Unexpectedly, it has been discovered that when the pump was focused inside the sample the spatial isotropy of the blue luminescence can be broken [Fig. 4(b)]: The luminescence scattering increases along the direction of the pump polarization, while the circular shape of the pump beam remains unchanged. If we rotate the direction of the pump polarization by using a half-wave plate, the elongated pattern of the blue luminescence follows this rotation [Figs. 4(c)–4(e)]. It should be noted that the blue luminescence was not polarized and self-focusing was not observed at peak powers used in the experiments. We called this phenomenon "propeller effect" due to the propellerlike shape of the luminescence spot in the focus of the pump beam. *The observed phenomenon represents the first evidence of anisotropic light scattering which peaks in the plane of light polarization in isotropic media.*

How can we explain this phenomenon? First, let us estimate the size of a light spot ($1/e^2$ intensity diameter) which is produced by the isotropically emitted luminescence in the focal plane of the microscope objective. Assuming that the luminescence is excited by the three-photon absorption of the pump at wavelength $\lambda = 800$ nm in a Gaussian beam with radius $r_0 = 2.3$ μm or by the one-photon absorption of UV (267 nm) third harmonic of the pump and that it is emitted isotropically all along the length of a beam waist, the size of the light spot a can be estimated as

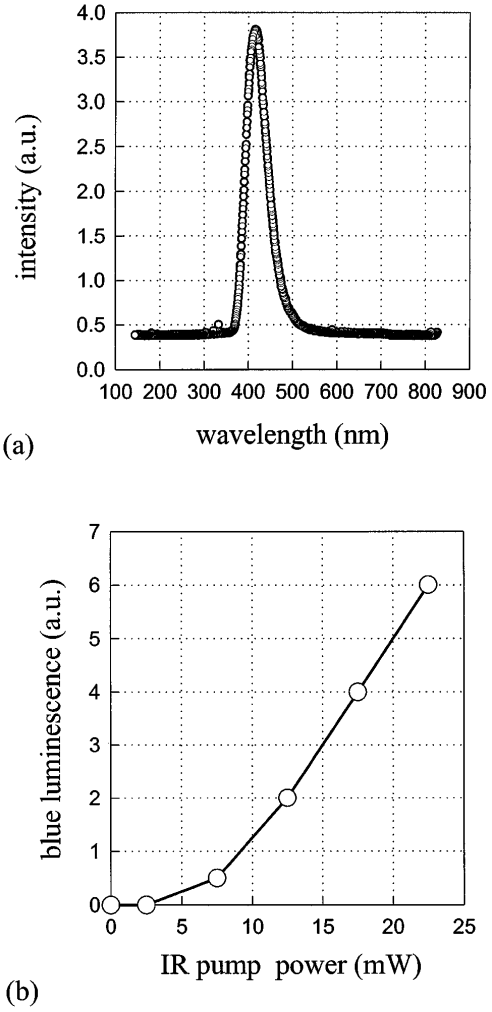

FIG. 2. Spectrum (a) and pump power dependence (b) of the blue luminescence.

$a \leq \pi r_0^2 n/\lambda = 30$ μm, where $n = 1.45$ is the refractive index of silica glass. This estimate is in a good agreement with the transverse size of the blue propeller [Figs. 4(b)–4(e)], which could be justified by ordinary (isotropic) luminescence. However, the longitudinal size of the blue propeller ($\sim$100 μm) is about 4.5 times larger than the transverse size of the propeller (Fig. 5). The fact that the blue luminescence is elongated along the pump polarization indicates that some additional momentum is acquired by the photons along this direction. We believe that such transformation of the momentum can be caused by the photoelectrons moving along the direction of pump polarization. The photoelectrons with the anisotropic momentum distribution can be created via the multiphoton ionization of defects (two-photon ionization of Ge-ODC from the long-lived triplet state) by the linearly polarized light of high intensity (Fig. 3). Indeed, it is well known that the angular distribution of photoelectrons can be elongated along the direction of the light polarization: $d\sigma/d\Omega \propto 1 + \cos^2 \theta$, where $d\sigma/d\Omega$ is the differential scattering cross section

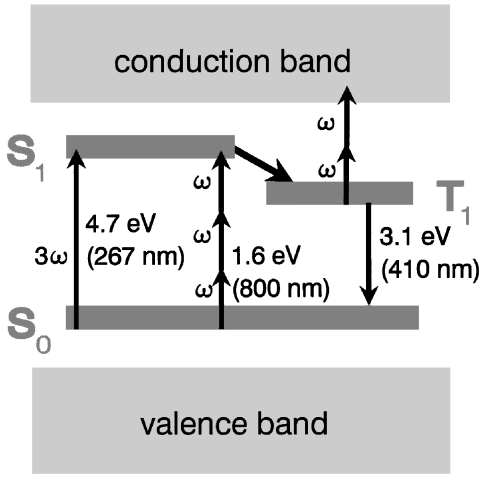

FIG. 3. Energy-level diagram of Ge-Si defect in silica glass with the possible channels of excitation.

of electrons and θ is the angle between the field amplitude vector $\mathbf{E}$ and the electron momentum vector $\mathbf{k}_e$ [22,23]. Assuming that the momentum relaxation time is about 100 fs and the speed of photoelectrons is 10^7 cm/s (which can be easily reached in the process of photoionization), the distance of a photoelectron path along the direction of the light polarization is about 10 nm. The distance between defects responsible for the blue luminescence (Ge-ODC at concentration of 10^{19} cm^{-3}) in the glass samples is $\sim$10 nm which is of the same order as the free path length estimated above. It is clear that the photoelectrons can be involved in a microscopic movement (with displacement of order of 10 nm) along the direction of the light polarization. Microscopic (much less than a wavelength of light) displacements of the photoelectrons along the direction of light polarization can lead to the anisotropic fluctuations of dielectric constant. Such fluctuations are obviously stronger along the direction of light polarization (in the direction of electron movement) compared to the perpendicular direction. The fluctuations of dielectric constant along the direction of light polarization induce index inhomogeneities which are elongated in the direction perpendicular to the pump polarization and which have $\mathbf{k}$ vectors of spacial harmonics parallel to the direction of polarization. The anisotropic inhomogeneities scatter photons (e.g., the ultraviolet photons of the third harmonic of the pump) in the plane of light polarization. Considering the angle of scattering $\varphi = 80°$ ($\tan\varphi = 3b/2z_0$, where $b = 100$ μm is the longitudinal size of the "blue propeller," $2z_0 = 2\pi r_0^2 n/\lambda = 60$ μm is the waist length of a pump beam), the size of these inhomogeneities can be estimated as $d \leq \lambda_{\mathrm{UV}}/(2n \sin\varphi) =$ 90 nm, where $\lambda_{\mathrm{UV}} = 267$ nm.

It should be pointed out that the scattering phenomenon described above must have strong wavelength dependence (λ^{-4}), which is similar to the wavelength dependence of Rayleigh scattering of light. Rayleigh scattering is

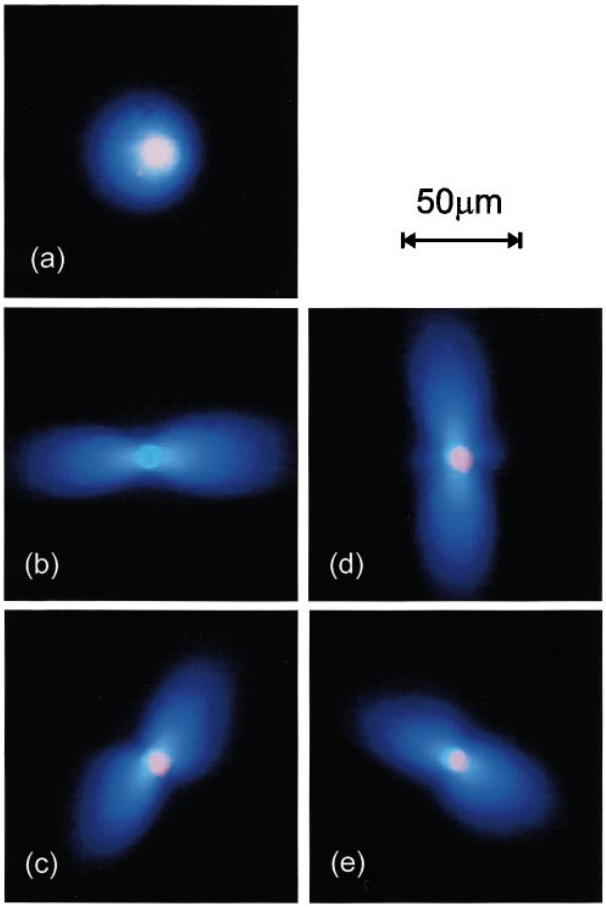

FIG. 4(color). Photographs of the blue luminescence spots in the focus of the linearly polarized pump. Pump is focused above the sample (a). Pump with four different orientations of polarization is focused inside the sample [(b)–(e)]. Notice that the blue luminescence spot is elongated along the pump polarization. The pink spot is produced by the pump leaking through the dichroic mirror.

normally caused by isotropic density fluctuations and the anisotropy in the scattering (the scattering is stronger in the direction perpendicular to the light polarization) is explained by the fact that a dipole does not radiate along its axis. In contrast to Rayleigh scattering the anisotropy in the observed scattering is caused by the anisotropy of the fluctuations itself. The strong dependence of the scattering on the wavelength can explain the absence of noticeable changes in the shape of the infrared pump.

Alternatively the photoelectrons involved in the movement along the direction of light polarization can directly transfer their momentum to the UV photons, e.g., via a Compton-like scattering. The electron momentum at a speed of 10^7 cm/s is about 50 times larger than the UV photon momentum. Even partial transfer of the electron momentum to the photons can create significant anisotropy in the light scattering. The photons, scattered indirectly (via the $\mathbf{k}$ vectors of the inhomogenieties) or directly by the photoelectrons, excite the anisotropic patterns of blue luminescence.

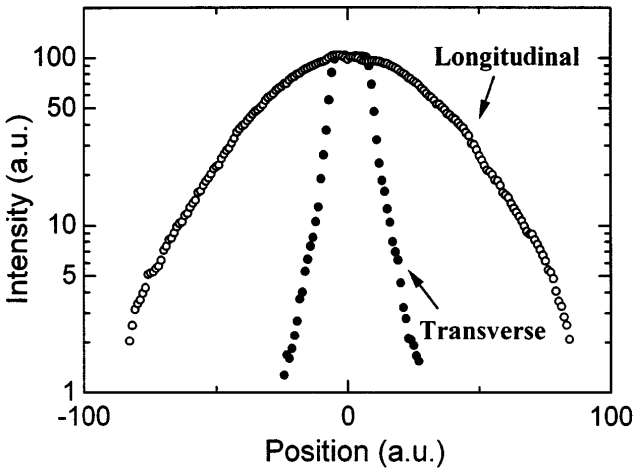

FIG. 5. Intensity distributions along the long axis of the "blue propeller" and in the perpendicular direction.

In other words, the pattern of the angular distribution of photoelectrons is imaged by the pattern of blue luminescence. To support our explanation we would like to point out a striking similarity of the observed patterns of the blue luminescence and the patterns of the angular distribution of photoelectrons in the experiments on the above threshold ionization (ATI) of atoms [22]. Indeed, the angular distribution of photoelectrons elongated along the light polarization has been observed in many experiments on atoms and the results reported in this Letter can be interpreted as the first evidence of such distribution in glass. Until now we have observed the phenomenon only in Ge-doped silica glass and experiments in other glass materials are in progress.

*Email address: pgk@orc.soton.ac.uk

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