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Memorized polarization-dependent light scattering in rare-earth-ion-doped glass

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We report the observation of memorized polarization-dependent light scattering in a Eu^{2+}-doped fluoroaluminate glass sample. Anisotropic light scattering along the plane of the light polarization was observed in the glass sample after the excitation of a focused 800 nm, 150 fs laser beam at a repetition rate of 200 kHz. When we changed the direction of the light polarization and irradiated the same location, we observed an anisotropic light-scattering pattern identical to the original one at the beginning, but then observed a new pattern along the new direction of the light polarization while the original light-scattering pattern disappeared gradually with the passage of time. This observed phenomenon was considered to be due to the light scattering of the polarization-dependent permanent microstructure induced by the polarized ultrashort pulsed laser itself. © *2000 American Institute of Physics.* [S0003-6951(00)01139-6]

The structure of glass is optically isotropic on a spatial scale larger than the wavelength of visible light. This is one of the important properties of glass in practical applications. However, a variety of light-induced anisotropic phenomena have been observed in recent decades.[1–8] In 1985, Parent *et al.* observed that reflecting Bragg filters having some polarizing properties are photoinduced in monomode optical fibers exposed to a linear polarized laser beam.[1] The reflectivity was found to reach a maximum when the polarization direction of a reading beam was parallel to the polarization direction of the writing beam. Since then, many photoinduced anisotropic phenomena have been observed and discussed for various types of glass.[2–8] Recently, Kazanski *et al.* observed an anisotropic light-scattering phenomenon that peaked in the plane of the light polarization when a Ge-doped glass was pumped by intense laser radiation.[8] The observed phenomenon was momentary. When they rotated the direction of the pump polarization, the elongated pattern of the blue luminescence followed the rotation. The phenomenon was considered to be due to the angular distribution of photoelectrons in isotropic solid materials.

In this letter, we report a memorized polarization-dependent light-scattering phenomenon in a Eu^{2+}-doped fluoroaluminate glass sample. The observed phenomenon is useful in the fabrication of polarization accessible optical memories, waveguide-type sensors, and other polarization-related micro-optic devices.

The composition of the sample used in the study was $1EuF_2 \cdot 14YF_3 \cdot 10MgF_2 \cdot 20CaF_2 \cdot 10SrF_2 \cdot 10BaF_2 \cdot 35AlF_3$ (mol %). 5N-purity grade EuF_3, YF_3, MgF_2, CaF_2, SrF_2, BaF_2, and AlF_3 were used as raw materials. A batch of 30 g was mixed in a glovebox filled with Ar gas. In order to convert residual oxides into fluorides, the batch was first melted in a glassy carbon crucible at 1100 °C under $Ar+NF_3$ (5 vol %) atmosphere for 1 h. The liquid was cooled to room temperature. The obtained glass was then put into a carbon crucible, and treated under an $Ar+H_2$ (5 vol %) atmosphere at 1100 °C for 1 h in a horizontal carbon furnace, while the reducing gas flowed from the top to the bottom of the furnace at a flow rate of 5 l/min. The specimen was obtained by cooling the melt at room temperature. The obtained glass specimen was cut and polished for the measurement of optical properties. The absorption spectrum of the sample was measured with a spectrophotometer (JASCO V-570). The photoluminescence was measured with a fluorescence spectrophotometer (SPEX 270M).

A regeneratively amplified 800 nm Ti:sapphire laser emitting 120 fs, 200 kHz, mode-locked pulses was used in our experiments. The laser beam (in the Gaussian mode at an average power of 100 mW) was focused using a 10× objective lens with a numerical aperture of 0.30 towards the interior of the glass sample of 4 mm thickness with an *XYZ* stage. The pump spot size in the focus of the beam was 8 μm. Simultaneously, the irradiated spot was imaged in the visible spectral range via an optical microscope using a color charge-coupled-device (CCD) camera as described in the previous paper.[8]

Figure 1 shows the absorption spectrum of the glass sample. No absorption peak in the wavelength region from 380 nm to 2.5 μm was observed. A broad peak can be observed at 250 nm, ranging from 190 to 380 nm. The peak can be assigned to the absorption of the $5d-4f$ transition of Eu^{2+}.[9] No apparent absorption due to the $4f-4f$ transition of Eu^{3+} was observed at 394 nm.[9,10] Therefore, most of the Eu ions existed as a divalent state in the glass.

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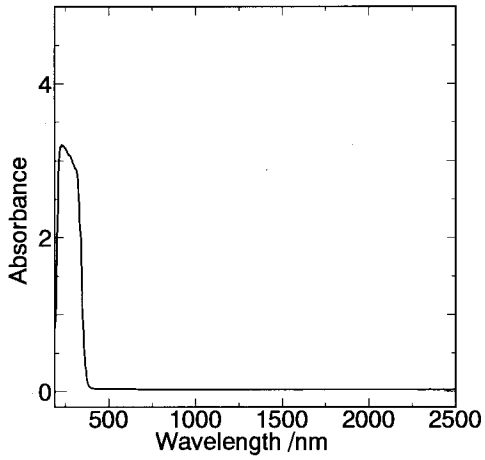

FIG. 1. Absorption spectrum of Eu^{2+}-doped fluoroaluminate glass.

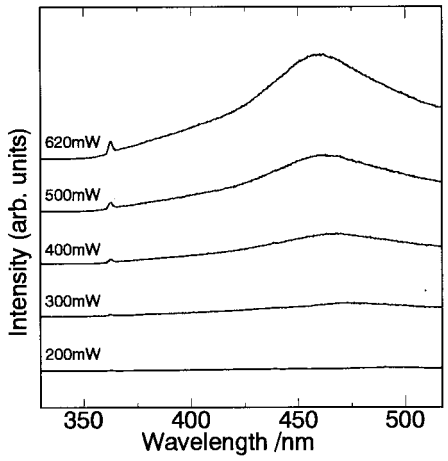

FIG. 3. Photoluminescence spectra of Eu^{2+}-doped fluoroaluminate glass excited by a femtosecond laser beam.

When the laser beam (0.5 μJ energy, 0.84 $\times 10^{12}$ W/cm^2 intensity in the focus of the beam) was focused slightly inside the sample, we observed a circular spot of blue luminescence via the observation of the optical microscope and CCD camera at the beginning. Then, a blue luminescence was observed along the direction of the pump polarization and the length of the blue-luminescence pattern increased with the passage of time. The pattern became stable after about 5 min; It is shown on the left-hand side of Fig. 2. Once formed, the anisotropic blue-luminescence pattern appeared instantaneously every time the laser beam's path to the sample was blocked and then unblocked. When we changed the direction of the light polarization and irradiated the same location, as shown in Fig. 2, we observed an anisotropic blue-luminescence pattern identical to the original one at the beginning, but then observed a new pattern along the new direction of the light polarization while the original blue-luminescence pattern disappeared gradually with the passage of time.

Figure 3 shows the photoluminescence spectra of the glass sample when excited by a femtosecond laser beam us-

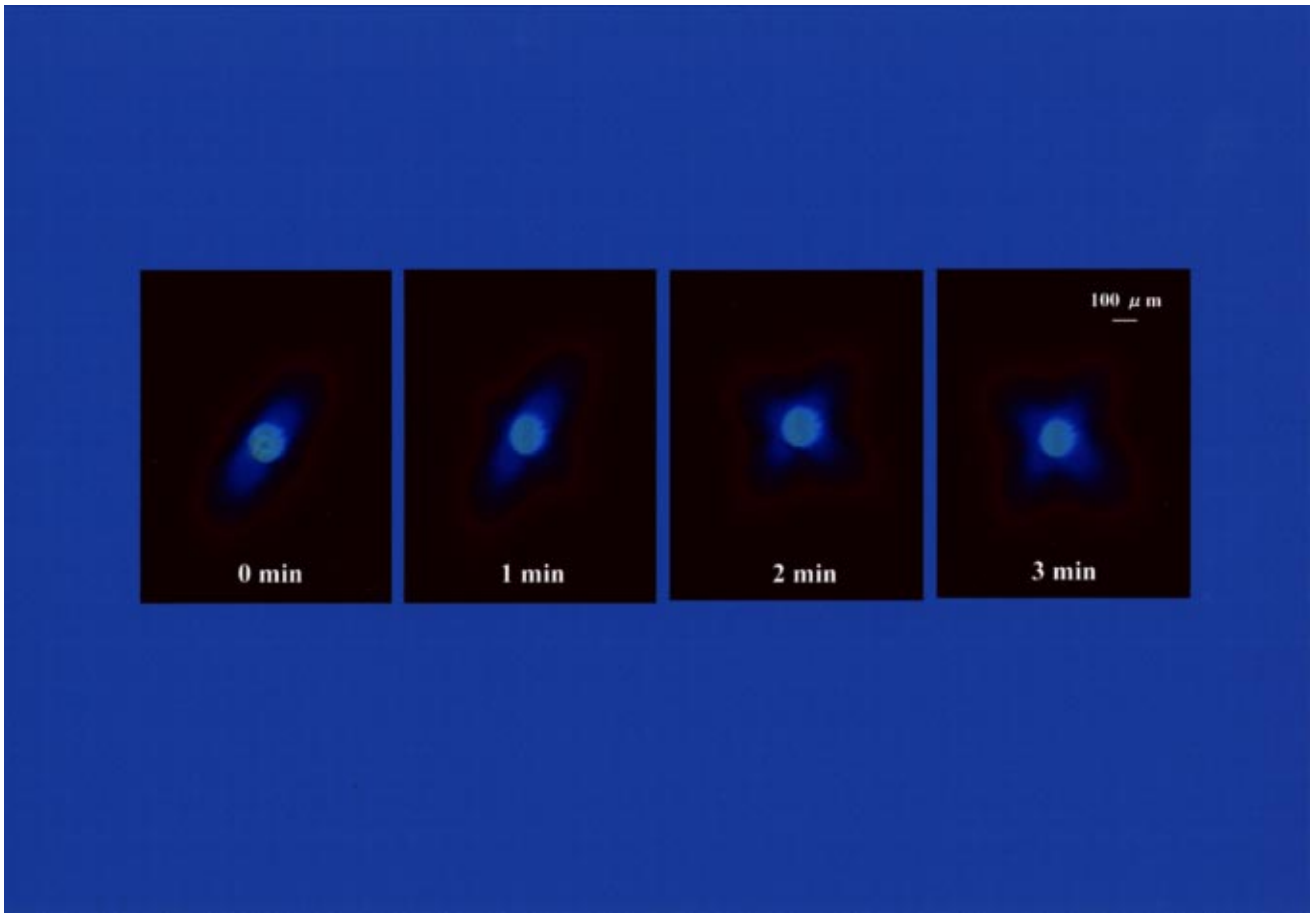

FIG. 2. (Color) Photographs of blue-luminescence patterns near the focus of a linear-polarized laser beam. The time shown in the figure is the duration after changing the polarization direction of the laser beam and irradiating the same location that was irradiated using the previous polarized laser beam.

ing a focusing lens ($f=100$ mm). A small peak exists at 360 nm due to the $^6P_{7/2}-^8S_{7/2}$ transition of Eu^{2+} ions and a broad peak exists at 460 nm due to the $5d-4f$ transition of Eu^{2+} ions.[10] Calculations showed that the intensity of the blue luminescence at 460 nm was nearly proportional to the three units of power of the excitation power of the femtosecond laser. Therefore, the blue luminescence at 460 nm was due to three-photon absorption and subsequent relaxation from the $5d$ level to the $^8S_{7/2}$ ground state of the Eu^{2+} ions.

What is the mechanism for permanent anisotropic luminescence in glass?

If the luminescence is excited by three-photon absorption of the pump at the wavelength, $\lambda=800$ nm and emitted isotropically along the length of the beam waist, the size of the light spot is about 100 μm. This is in agreement with the size of the circle spot at the center. However, the longitude of the blue luminescence is three times longer than the transverse. Therefore, the anisotropic blue luminescence along the direction of the light polarization is caused by some additional momentum, and is a light-scattering phenomenon as observed in Ge-doped silica glass.

It is typical for luminescence in glass to be isotropic and insensitive to the polarization of the excitation light due to the isotropic structure of glass. The anisotropic light-scattering phenomenon in Ge-doped silica glass has been explained by anisotropic index fluctuations excited by electrons moving along the direction of the light polarization in the process of photoionization by intense light. These fluctuations scatter strongly in the plane of the light polarization for short-wavelength light (similar to Rayleigh scattering), e.g., ultraviolet light generated in glass. The ultraviolet light is absorbed by defects in the Ge-doped silica glass and excites the anisotropic pattern of luminescence.[8]

In the present case, the anisotropic light-scattering pattern was permanent and could be modified with another polarized beam. Therefore, the anisotropic light scattering in the Ge-doped silica glass cannot be directly used to explain the observed phenomenon in the Eu^{2+}-doped glass. We also measured electron-spin-resonance spectra in both types of glass under the same laser-irradiation conditions, and observed signals due to the permanently formed electron and hole trapping centers in them. Therefore, we suggest that the polarization-dependent permanent structure was induced during the intense laser irradiation.

Ouellette, Gagnon, and Poirier observed an anisotropic permanent refractive-index change in Ge-doped fibers exposed to mode-locked light at 532 nm.[2] The refractive-index change followed the direction of the polarization of the writing light. We suggest that a similar permanent refractive-index change was induced in the Eu^{2+}-doped glass; only the permanent polarization-dependent structure was induced here via multiphoton processes.

The focused femtosecond laser beam not only induces electron and hole trapping centers in the glass via multiphoton absorption and multiphoton ionization, but also acts as a driving force for inducing the distribution of induced defects, since a pair of electron and hole centers can be considered as a dipole.[11] The driving force in the polarization direction is larger than that in the direction perpendicular to the light polarization; therefore, the permanent refractive-index fluctuations in the direction parallel to the light polarization are larger than those in the direction perpendicular to the light polarization. This structure results in stronger light scattering, e.g., Rayleigh scattering, in the direction of the light polarization because the scattering is proportional to the density or refractive-index fluctuations. The pump beam is scattered by the pump-beam-induced polarization-dependent permanent structure, and the scattered light excites the $^8S_{7/2}$ ground state to the $5d$ level of Eu^{2+} via three-photon absorption, finally resulting in the anisotropic blue luminescence.

When the same location is irradiated in another direction of the light polarization, the previously induced structure is destroyed and a new polarization-dependent structure is created. Therefore, a blue-luminescence pattern can be observed along the new direction of the light polarization. Since this observed phenomenon is related to the permanent polarization-dependent structure, we expect that it is possible to fabricate polarization accessible three-dimensional optical-memory and waveguide-type micro-optic devices.[12,13]

In conclusion, we have observed memorized anisotropic light scattering in a Eu^{2+}-doped fluoroaluminate glass sample. Anisotropic blue luminescence, which peaked in the plane of the light polarization was observed in the glass sample pumped by a focused 800 nm, 150 fs, laser light at a repetition rate of 200 kHz. When we changed the direction of the light polarization and irradiated the same location, we observed an anisotropic blue-luminescence pattern identical to the original one at the beginning, but then observed a new pattern along the new direction of the light polarization while the original light-scattering pattern disappeared gradually with the passage of time. This observed phenomenon was considered to be due to the light scattering of the polarization-dependent permanent microstructure induced by the polarized ultrashort pulsed laser itself.

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