

Anomalous photoluminescence polarization of porous silicon

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A linear polarization of the photoluminescence of porous silicon has been observed during high-energy nonresonant excitation by linearly polarized light. The depolarization time of this luminescence is anomalously long ($\sim 10 \mu\text{s}$) at temperatures up to $T = 300 \text{ K}$.

The discovery of an efficient visible photoluminescence of porous silicon¹ has triggered active research on this material and on the mechanism for its photoluminescence. A linear polarization of the photoluminescence in porous silicon has been observed² at low temperatures during excitation into the photoluminescence band by the beam from an argon laser. Observation of two emission bands in time-resolved photoluminescence spectra of porous silicon was reported in Ref. 3. These were a short-wavelength band *A1*, with a nanosecond lifetime, and a long-wavelength band *A2* with a lifetime of $10 \mu\text{s}$. The latter band determines the luminescence spectrum during steady-state photoexcitation.

In the present letter we are reporting the observation of a linear polarization of the photoluminescence of porous silicon during high-energy nonresonant excitation by linearly polarized light under conditions such that there is a significant energy loss, $\sim 1\text{--}2 \text{ eV}$. Study of the time evolution and spectra of the photoluminescence has revealed the contributions of various emission channels to the shaping of the time-resolved and steady-state spectra of the degree of linear polarization ρ of the emission of porous silicon. We studied layers of porous silicon in the (100) orientation, prepared by the procedure of Ref. 3. The luminescence was excited by a linearly polarized beam from a pulsed N_2 laser ($t_p = 10 \text{ ns}$, $\lambda = 3370 \text{ \AA}$, $P = 10^2\text{--}10^3 \text{ W/cm}^2$) and also by cw beams from an Ar^+ laser ($\lambda = 4880 \text{ \AA}$) and a He-Ne laser ($\lambda = 6328 \text{ \AA}$) with $P \sim 1 \text{ W/cm}^2$. The exciting light was incident normally on the surface of the crystal; the photoluminescence was observed at a small angle from the propagation direction of the exciting light. The photoluminescence was measured with a DFS-24 double monochromator, a pulsed measurement system with a time resolution $\sim 20 \text{ ns}$, and a photon counting system.

Figure 1a shows time-resolved spectra of the photoluminescence. There are two bands,³ *A1* and *A2*. The photoluminescence is linearly polarized over the entire wavelength range. The value of the degree of linear polarization,⁴ $\rho = (I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})$, reaches 0.3 at the violet edge of the *A1* band (Fig. 1b). Here I_{\parallel} is the intensity of the photoluminescence polarized in the same way as the exciting light, while I_{\perp} is the intensity of the photoluminescence polarized in the perpendicular direction. When the measurement of the photoluminescence is taken $1 \mu\text{s}$ after the

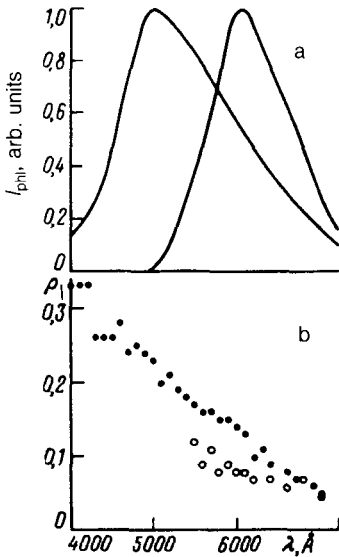


FIG. 1. a: Time-resolved photoluminescence spectra of porous silicon at $T=300$ K at various times after the beginning of the laser pulse, Δt . 1— $\Delta t=10$ ns; 2— $1 \mu\text{s}$. b: Spectra of ρ of the time-resolved spectra. ●— $\Delta t=10$ ns; ○— $1 \mu\text{s}$.

peaking of the laser pulse, in the case in which the time-resolved spectrum is formed by band A2 alone, the value of ρ for this band is 0.06–0.1. At a delay of 10–100 ns, for which the time-resolved spectra are formed by a superposition of A1 and A2, the spectrum of ρ has contributions from both bands. The degree of linear polarization does not depend on the orientation of the pump polarization in the plane of the test sample; the two polarizations are parallel. The relaxation time for ρ of the emission at the peak of band A2 (Fig. 2) is a few microseconds ($\sim 10 \mu\text{s}$), on the order of the luminescence lifetime. Figure 3 shows the ρ spectrum of the photoluminescence for various energies of the exciting photon. A decrease in the difference between the

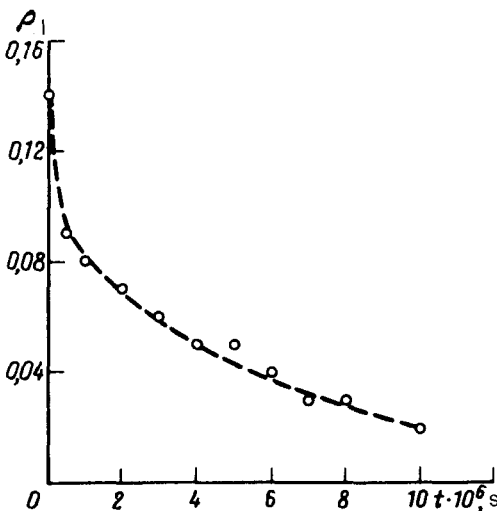


FIG. 2. Kinetics of ρ of the photoluminescence of porous silicon during pulsed laser excitation at $T=300$ K with $\lambda_{\text{meas}}=6500 \text{ \AA}$.

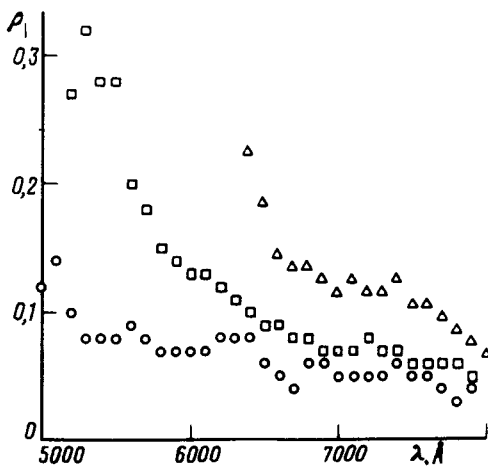


FIG. 3. Spectra of ρ of the photoluminescence of porous silicon at $T=300$ K for various wavelengths of the exciting light. \circ — $\lambda=3370$ Å (quasi-steady spectrum); \square — $\lambda=4880$ Å; \triangle — 6328 Å.

energies of the exciting photon and the photoluminescence photon leads to an increase in the degree of polarization of the photoluminescence. We also observe a decrease in the degree of polarization in the long-wavelength wing of the photoluminescence band. It was found that a magnetic field up to 36 kOe in the Faraday geometry has essentially no effect on ρ .

A retention of the linear polarization of the exciting light in the emission from semiconductors has been observed previously, e.g., for the hot photoluminescence of direct-gap semiconductors.⁴ This effect is attributed to an alignment of the momenta of photoexcited charge carriers by linearly polarized light. However, this alignment is observed only near the pump spectral line, and it disappears after several phonons have been emitted by the nonequilibrium carriers. The time scale over which the nonequilibrium distribution function reaches isotropy and thus the relaxation time of the linear polarization of the photoluminescence are 10^{-13} – 10^{11} s under these conditions. The decay time of the linear polarization of the photoluminescence which we observed is much longer, implying a completely different mechanism for the linear polarization of the photoluminescence in porous silicon. This conclusion is also implied by the ρ spectrum. The polarization is preserved when the difference between $\hbar\omega_{\text{exc}}$ and $\hbar\omega_{\text{phl}}$ is 1–2 eV (Figs. 1 and 3). This difference corresponds to the energy of 20 to 40 optical phonons. The decrease in ρ with increasing $\hbar\omega_{\text{exc}}$ and also with increasing difference between $\hbar\omega_{\text{exc}}$ and $\hbar\omega_{\text{phl}}$ (Fig. 3) is evidence that the energy relaxation has a comparatively slight effect on the degree of linear polarization.

We believe that the photoluminescence polarization observed here can be understood under the assumption that this system is a set of randomly distributed 1D nanocrystals which cause an efficient visible photoluminescence.

The absorption of linearly polarized light should be anisotropic in each individual nanocrystal because the latter is aspherical. In the case of a quasi-1D nanocrystal, whose long dimension is parallel to the polarization of the exciting light, the absorption is much stronger than that for a nanocrystal which is oriented, say, perpendicular to the polarization of the exciting light. It can be shown that a quasi-1D geometry

should also cause the thermalized luminescence to be anisotropic in the polarization sense. This luminescence will be polarized predominantly along the direction of the long dimension of the nanocrystal. The ratio of the length of nanocrystal, L , to its width D , i.e., $\sim L/D$, must be a parameter of this anisotropy. A random distribution of nanocrystals will lead to a situation in which the number of quasi-1D nanocrystals, which are preferentially excited, will be essentially the same for any direction of the vector E of the exciting light. The emission from the entire set of excited nanocrystals will be polarized linearly along the direction of the pump polarization, and the polarization vector of the photoluminescence will track the direction of the polarization vector of the exciting light as it rotates.

If there is no migration of energy between nanocrystals, the photoluminescence will be linearly polarized as long as it exists. If there is an energy migration, the photoluminescence will be depolarized. This model easily explains why a magnetic field has no effect on the degree of linear polarization of the photoluminescence.

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