Proof of the existence of optical anisotropy in the EL2 centers of gallium arsenide by the polarized-luminescence method

E. I. Oborina, S. S. Ostapenko, and M. K. Sheĭnkman
Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR

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The optical anisotropy of the 0.635-eV and 0.68-eV photoluminescence centers of the EL2 series has been established by the polarized-luminescence method. Each band is shown to be related to two defects with different symmetries, one of which is unstable. A persistent relaxation of the induced polarization of the photoluminescence accompanying the fatiguing process has been detected for the first time.

The photoluminescence bands with peaks at 0.635 eV (L_1) and 0.68 eV (L_2) in gallium arsenide are usually related to the intrinsic defects which belong to the EL2 series and which account for the semiinsulating properties of the material.[1] Although the basic recombination characteristics of these centers have been studied in detail, their physicochemical nature and their structure have not been definitively established. To solve this problem, it is necessary to determine the symmetry and optical anisotropy of the defects. The data available, however, are contradictory. While the anisotropy of the EL2 defects has been established on the basis of the capacitive changes,[2] the piezospectroscopic studies of the intracenter absorption by these defects have shown that the center has a T_d symmetry.[3]

To study the optical anisotropy of the defects responsible for the L_1 and L_2 luminescence bands, we have used for the first time the polarization spectroscopy of the photoluminescence and of its optical excitation. To use this method, the optical excitation spectrum must have a band, which is associated with the direct excitation of the luminescence center or with the indirect excitation, in which the neighboring center is excited, followed by a directional (anisotropic) transfer of energy to the luminescence center.

The maximum of the excitation of the 0.87-eV (E_3) luminescence band L_1 (curve 1 in Fig. 1) is associated with the direct excitation of the center, which stems from the Stokes shift $\Delta_s = 110 \pm 10$ meV (Ref. 1). We see that the optical excitation spectrum of this photoluminescence band also has E_1 (1.47 eV) and E_2 (1.35 eV) maxima. The optical excitation spectrum of the L_2 band is identical. The E_1 band was interpreted by Paget and Klein[4] as an optical transition from the V zone to the shallow donor. The excitation maximum in the E_2 band, which was observed by Samuelson *et al.*,[5] will be discussed in another paper.

Excitation of the two photoluminescence bands, L_1 and L_2, by light in the interval $L\nu_E$ = (1.0–1.35 eV) causes the photoluminescence intensity to decay with time from the initial value I_i to the steady-state value I_{ss} in approximately exponential manner

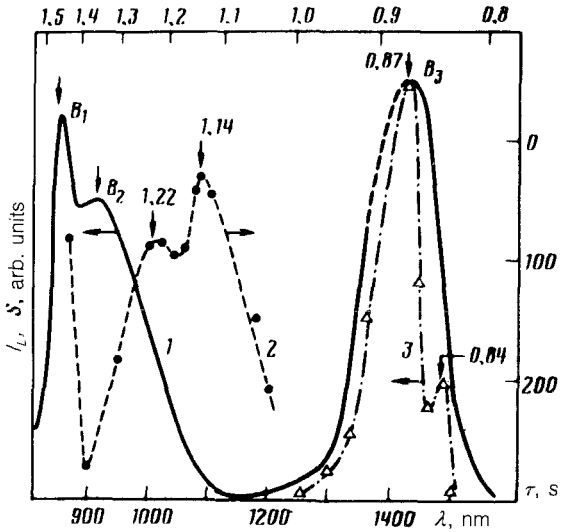

FIG. 1. 1—Spectrum of the excitation of the L_1 band ($h\nu_L = 0.58$ eV) at 77 K; 2—spectrum of the characteristic photoluminescence fatiguing time; 3—spectrum of the restoration of photoluminescence; S—light sum restored in a time $\Delta t = 2$ min.

$\exp(-t/\tau)$ (Fig. 2a). This "fatiguing" of the photoluminescence, which is characteristic for the EL2 defects, is associated with the existence in them of a metastable state.[1] The spectral dependence of τ^{-1}, which is represented by curve 2 in Fig. 1, reveals the presence of 1.14-eV and 1.22-eV maxima, in satisfactory agreement with the calculated energies of the optical transitions between the ground state and the excited state of the EL2 defects.[6]

If the sample is illuminated by light from the E_3 band at the same temperature

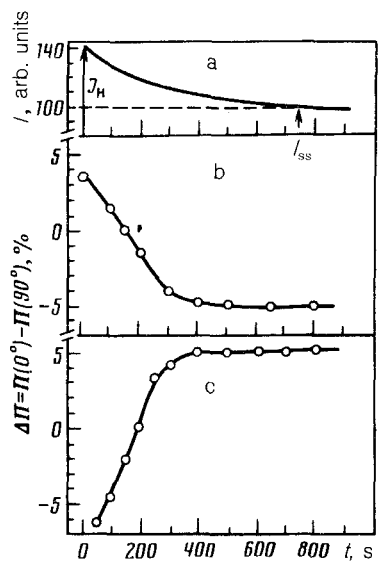

FIG. 2. Photoluminescence kinetics during the fatiguing process ($h\nu_L = 0.58$ eV); b—kinetics of the induced polarization of the L_1 band ($h\nu_L = 0.58$ eV); c—kinetics of the induced polarization of the L_2 band ($h\nu_L = 0.7$ eV). The exciting light is incident normal to the (001) plane; the luminescence is detected in the (110) plane ($h\nu_E = 1.32$ eV). The choice of $h\nu_E$ in the region of slow photoluminescence kinetics is stipulated by the method used to measure $\Delta\Pi(t)$, $T = 77$ K.

(77 K) after the I_{ss} is reached, the photoluminescence intensity will recover to the value I_i. The spectrum for the recovery of the luminescence light sum has peaks at 0.84 eV and 0.87 eV (curve 3 Fig. 1). Optical recovery of luminescence by extrinsic light, which stems from the inverse transition of the defect from the metastable state to the ground state, to the best of our knowledge, has heretofore not been observed.

We used the polarized-luminescence method to excite the luminescence of L_1 and L_2 in the E_2 and E_3 bands. The principle upon which this method[7] is based is the measurement and analysis of the so-called polarization diagram:

$$\Pi(\varphi) = \frac{J_{\parallel}(\varphi) - J_{\perp}(\varphi)}{J_{\parallel}(\varphi) + J_{\perp}(\varphi)}, \quad (1)$$

where $J_{\parallel}(\varphi)$ and $J_{\perp}(\varphi)$ are the angular dependences of the intensities of two photoluminescence fluxes polarized orthogonally to the principal crystallographic direction (the $\langle 110 \rangle$ direction, in our case), and φ is the angle between the electric vector of the exciting light and the $\langle 110 \rangle$ axis. The governing parameter is the induced polarization of light $\Delta\pi = \pi(0°) - \Pi(90°)$. The satisfaction of the inequality $\Delta\pi \neq 0$ is the criterion of the optical anisotropy of the luminescence center, i.e., of its low local symmetry.

We found that $\Delta\pi \neq 0$ in each photoluminescence band (L_1 and L_2) when they are excited in the E_3 band and that $\Delta\Pi$ does not change over time. Each type of photoluminescence center is therefore optically anisotropic. We also found that $\Delta\pi \neq 0$ upon the excitation of the two photoluminescence bands in the interval $h\nu_E = (1.0\text{–}1.35)$ eV, but the induced polarization now changes as a function of time in accordance with the behavior peculiar to each photoluminescence band (Fig. 2, b and c). These functional dependences $\Delta\Pi(t)$, which were observed for the first time and which in the L_1 band are different from those in the L_2 band, are proof that the optical anisotropy is different in the corresponding defects and that the induced polarization changes as a function of time. The presence of such functional dependences can be attributed to either the transition of all photoluminescence centers to a new metastable state, which is characterized by a lower quantum radiation yield and a distinct optical symmetry, or to the presence in each band (L_1 and L_2) of stable defects and relaxation defects, whose luminescence spectra are virtually indistinguishable but whose values of $\Delta\Pi$ are distinct. Quantitative analysis of the $\Delta\Pi(t)$ curves (Fig. 2, b and c) showed that the second possibility occurs in our case. The value of $\Delta\Pi$, calculated for the changing part of the intensities of the photoluminescence bands, remains constant during the fatiguing process. This value ranges between $+(14 \pm 2)\%$ and $-(12 \pm 2)\%$, respectively.

We have thus shown that each 0.635-eV and 0.68-eV photoluminescence band is described by two types of defects which are distinguished by optical symmetry and stability. The fatiguing process occurs as a result of the decrease in the concentration of unstable centers with a high degree of optical anisotropy.

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