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EXPERIMENTAL EVIDENCE FOR THE SPATIAL CORRELATION OF CHARGED EL2(+) DEFECTS AND ACCEPTORS IN SEMI-INSULATING GaAs*

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The far-infrared photoconductivity due to shallow donors was measured in semi-insulating GaAs for different states of the EL2 defect — normal, metastable, and during the transition — with and without hydrostatic pressure. The results show that the intra-donor transition line broadening observed previously after transferring the EL2 to the metastable configuration cannot be due to lattice distortion effects, and can only be explained in terms of a spatial correlation of charged EL2(+) and acceptor A(-) states, which form electric dipoles.

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In semi-insulating (SI) GaAs the concentration of free electrons in the conduction band (CB) and that of neutral shallow donors at thermal equilibrium at 4.2 K is practically zero. The Fermi energy is pinned at the EL2 level, an intrinsic defect forming a deep donor located 0.75 eV below the CB edge [1]. In a simple 3-level compensation model [2], the concentrations of shallow donors, shallow acceptors and EL2 defects are such that

$$N_{\rm D} < N_{\rm A} < N_{\rm D} + N_{\rm EL2}.$$

At 4.2 K all shallow donors and acceptors, as well as $N_{\rm EL2+} = N_{\rm A} - N_{\rm D}$ EL2 defects are ionized. The total number of charges, positive and negative, when the EL2 is in the normal configuration, is

$$\sum Q_{\text{normal}} = N_{\text{D}+} + N_{\text{A}-} + N_{\text{EL}2+} = N_{\text{D}} + N_{\text{A}} + N_{\text{A}} - N_{\text{D}} = 2N_{\text{A}}.$$
 (2)

The large number of point charges (of the order of 10^{16} cm⁻³) and practically no free electrons to screen them, results in strong fluctuations of the conduction and valence bands, and thus also in fluctuations of the local electric fields.

Illumination with 1.05 μ m light transfers the EL2 into its metastable configuration, where it is optically and electrically inactive [3, 4]. This transition is accompanied by a significant lattice distortion [5]. Only EL2⁰ states undergo this

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transition. The bleaching process may be divided into two parts: the first during which $EL2^0$ states are transferred into the metastable state, $EL2^0 \rightarrow [EL2^0]^*$, and the number of charges remains unchanged, and the second, when ionized $EL2^+$ centres capture electrons, are neutralized and then transferred into the metastable configuration. This may be written as $EL2^++A^- \rightarrow A^0 + [EL2^0]^*$. Thus, after total bleaching, the number of charges is

$$\sum Q_{\text{metastable}} = N_{\text{D}+} + N_{\text{A}-} = N_{\text{D}} + N_{\text{D}} = 2N_{\text{D}}.$$
(3)

Note that, from Eq. (1), $\sum Q_{\text{metastable}} < \sum Q_{\text{normal}}$.

Under hydrostatic pressure the acceptor-like level of the EL2 [6, 7] enters the band gap and may be populated, i.e. a defect in the $[\text{EL2}^0]^*$ state captures an extra electron. No lattice relaxation accompanying this process was found. Furthermore, under hydrostatic pressure the EL2 can be transferred by means of light from the metastable back to the normal configuration — an equilibrium between the two processes, forward and reverse, is reached. The process of populating $[\text{EL2}^0]^*$ states by electrons may be schematically written as $\text{EL2}^0 + [\text{EL2}^0]^* \rightarrow \text{EL2}^+ + [\text{EL2}^-]^*$. If we denote the dynamic equilibrium between EL2 defects in the normal and metastable states by $\alpha = N_{\text{EL2}^*}/N_{\text{EL2}^0}$, we can write the total number of charges in the metastable configuration under pressure as

$$\sum Q_{\text{met.press}} = N_{\text{D+}} + N_{\text{A}} + N_{\text{EL2+}} + N_{\text{EL2-}}$$

$$= N_{\rm D} + N_{\rm A} + N_{\rm A} - N_{\rm D} + \alpha N_{\rm EL2^0} + \alpha N_{\rm EL2^0} = 2N_{\rm A} + 2\alpha N_{\rm EL2^0}.$$
 (4)

The number of charges is thus increased, although overall neutrality is of course conserved.

We have previously shown [8] that the magneto-spectroscopy of shallow donor states can be used to obtain information concerning local electric fields in a semiconductor. A theory of the linewidth of intra-donor transitions was developed by Larsen [9], who calculated the effect of the electric fields of point charges on the lineshape. The theory, basing on Stark-effect shifts of transition energies, was valid for ultrapure samples (ionized impurity concentrations $\approx 10^{14}$ cm⁻³). Above this limit lifetime broadening (instability of the excited state due to field ionization) became important. Later calculations [10] showed that the average electric field was reduced if spatial correlations between charges (dipoles) were taken into account. Typical SI GaAs represents a range of concentrations where existing theories are not able to give quantitative predictions of linewidths; however, it can be stated that linewidths increase with increasing electric fields and that spatial correlations decrease these fields.

The experimental setup was described previously [8]. Continuous illumination with light of wavelengths between 0.86 μ m and 1.05 μ m served to obtain a steady-state population of CB and shallow donor states. The photocurrent resulting from the additional excitation, by far-infrared (FIR) light, of such photoneutralized donors was measured with a lock-in amplifier synchronized with the chopped FIR radiation.

Figure 1 shows spectra taken before and after bleaching the EL2. When bleaching is performed under pressure, the linewidth can be seen to change dramatically, by about 100%. In comparison, the broadening caused by bleaching



Fig. 1. Tracings of photoconductivity spectra vs. magnetic field. The temperature was 4.2 K, the laser energy 118.8 μ m. Curve a — EL2 in the normal configuration, curve b — EL2 completely bleached, without pressure, curve c — EL2 bleached under hydrostatic pressure (0.4 GPa). The curves have been scaled to fit on one graph.

without pressure is only of the order of 50%. Since bleaching under pressure differs from bleaching without pressure by the additional (purely electronic) process of electron capture onto $[EL2^0]^*$ states, it can be seen that the dominant mechanism causing line broadening is connected with electric charges (compare Eqs. (3) and (4)), and not with lattice relaxation. Further proof is given in Fig. 2, which shows linewidths of spectra taken during bleaching: the sample was illuminated with 0.86 μ m light while each spectrum was recorded, and with 1.05 μ m (bleaching) light during 5-minute intervals between the spectra. The shape of the spectrum changed slightly, but the linewidth of the $1s-2p_+$ transition remained constant. The horizontal axis in Fig. 2 is only qualitative — there is no good quantitative measure of the number of EL2 defects which were transferred to the metastable configuration. It can only be stated that the number of defects in the normal configuration decreases from left to right. It can be seen that the change in linewidth occurs only during the second part of the bleaching process, i.e. when the number of charges changes.

These findings confirm previous suggestions [8] that, since the number of charges in the sample decreases with EL2 bleaching (see Eqs. (2) and (3)) while the linewidth increases, ionized EL2(+) defects must be spatially correlated with acceptors. Such dipoles would screen the fields of the remaining point charges, and the average electric fields before bleaching would then be smaller than after bleaching, when there are less charges. The fact, shown above, that the line broadening is explicitly connected with the number of charges excludes the possible role of lattice distortion, and thus leaves the dipole hypothesis as the only possible explanation of the observed phenomena.



Fig. 2. Linewidth of the $1s-2p_+$ transition as a function of EL2 transferred into the metastable configuration. The horizontal axis is purely qualitative — see text. Empty squares correspond to the first part of the bleaching process, when there is no change in the number of charges (0.86 μ m light used). The filled square on the far right represents the end of the process, when all EL2 defects are in the metastable state (0.63 μ m light used). The filled square on the left is for all defects in the normal configuration, with 0.63 μ m light. The slightly narrower line is probably due to ionization of acceptors with this pumping wavelength.

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