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Optical activity of the *EL2* metastable state under hydrostatic pressure

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We present experimental results showing that at hydrostatic pressure exceeding approximately 0.3 GPa the *EL2* metastable state becomes optically active in both *n*-type and semi-insulating GaAs, and the pressure-induced optical recovery (PIOR) can be easily observed. PIOR is practically full and extremely efficient (with an efficiency comparable with that of the photoquenching process). The proper choice of the energy of the illuminating light allows reversible cycles of photoquenching and subsequent optical recovery of the characteristic intra-*EL2* absorption at 5 K. PIOR is a few orders of magnitude more efficient than pure optical recovery processes already reported for atmospheric pressure.

It is well known that in GaAs the *EL2* defect in its neutral charge state manifests itself in the photoionization background starting from 0.77 eV and the charactertistic intra-*EL2* absorption band ranging from 1.04 to 1.4 eV.[1,2] At low temperature this absorption can be completely quenched by means of irradiation with photons from the intra-*EL2* absorption band.[3] This photoquenching is the optically induced transformation of the *EL2* defect (without change of its charge state[4,5]) from its normal to its metastable state. Both states are EPR inactive[6-8] and the latter is also believed to be optically inactive.[9] Without illumination the *EL2* absorption recovers if the sample temperature is raised above 40 or 130 K for *n*-type and semi-insulating (SI) samples, respectively.[10,11]

Because metastability is the key problem in the study of the nature of the *EL2* defect and the metastable state itself cannot be investigated directly, the process of *EL2* recovery is of crucial importance. Besides pure thermal recovery processes, mentioned above, there have also been reported so-called optically assisted thermal recovery processes of the *EL2* observed above 60 K (Ref. 12) as well as pure optical recovery effects[13-19] (the observation of the latter processes at low temperatures makes the optical inactivity of *EL2* metastable state questionable). Pure optical recovery effects were reported for absorption,[13,14] photoluminescence,[15,16] electron paramagnetic resonance[17] (where the concentration of the $EL2^+$ was monitored), photoconductivity,[18] and photocapacitance[19] experiments. The following should be pointed out.

(i) All these experiments were performed on SI samples[20] where the role of $EL2^+$ should be taken into account.

(ii) The reported recovery effects were only partial (the amplitude of the recovered absorption not exceeding 27% of the initial $EL2^0$ absorption[14]).

(iii) In most experiments high-intensity (laser) light was used to induce the recovery process.

(iv) The efficiency of the recovery process was always very low—the time constant of the recovery being 3 orders of magnitude larger than that of the photoquenching process [e.g., 23 min compared with 1.5 s (Ref. 14)].

(v) All the reported purely optical recovery processes took place for photon energies lower than the intra-$EL2^0$ absorption band.[20]

In the present paper we give experimental evidence that at hydrostatic pressure the *EL2* metastable state becomes optically active with an extremely efficient purely optical recovery process that is not observed at ambient pressure.

The experiment was performed on two kinds of crystals: SI liquid-encapsulated Czochralski (LEC) GaAs and horizontal Bridgman *n*-type GaAs:Si with the Hall concentration measured at 5 K equal to 1.2×10^{16} cm^{-3}. Samples were placed in a high-pressure optical cell with helium as a pressure-transmitting medium. Optical transmission spectra were measured at 5 K in the spectral range 0.55–1.4 eV with the use of a tungsten halogen lamp as a light source, a prism monochromator, and a PbS detector. The same optical setup was used to irradiate the samples and to monitor the photoquenching and optical recovery processes. Under high pressure we could not apply the standard in-out method and we normalized the recorded spectra with the use of the spectra taken without a sample in a separate experiment. Thus only the relative absorption coefficient $\Delta\alpha$ could be determined (unknown additive constant). During the measurements of the absorption spectra the light intensity was maintained a few orders of magnitude smaller than in the case of the optical quenching and recovery investigations. At each pressure the sample was cooled down in the dark starting from approximately 160 and 70 K for the SI and *n*-type sample, respectively. The whole experiment consisted of a series of the following irradiation cycles: (I) the absorption spectrum measurement; (II) monochromatic-light irradiation of the sample (approximately 15 min for each cycle) with the simultaneous monitoring of the time dependence of the transmission coefficient for the irradiating light; (III) the same as (I) to verify the influence of the irradiation. Figures 1 and 2 present some examples of the experimental spectra obtained during the above-mentioned irradiation cycles for both crystals investigated at high pressure. It is clear that at high pressure, after the optically induced transfer of the *EL2* to its metastable state (1.13-eV photoquenching) the *EL2* could recover to its fundamental state by means of the appropriate monochromatic light irradiation, and the initial absorption spectrum could be restored (see, e.g., spectrum *c* from Fig. 1 or spectrum *d* from Fig.

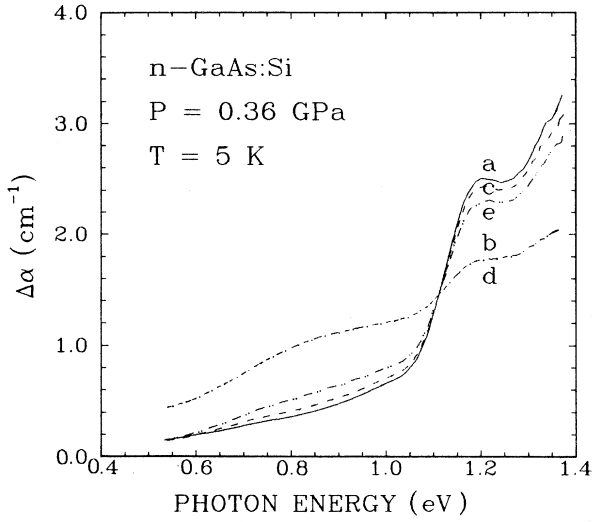

FIG. 1. The influence of the succeeding processes of the monochromatic light irradiation on the *EL2* absorption spectrum at 0.36 GPa for *n*-GaAs:Si at 5 K: (a) initial spectrum (after cooling the sample down), (b) after 1.13-eV irradiation, (c) after 1.36-eV irradiation, (d) the same as (b), and (e) after 1.0-eV irradiation.

2). This pressure-induced optical recovery (PIOR) appeared gradually at pressures 0.25–0.3 GPa, so that at $p > 0.3$ GPa the experimental results were qualitatively the same irrespective of pressure. The most important features of the PIOR effect were the following.

(1) PIOR existed in both *n*-type and SI GaAs at pressures $p > 0.3$ GPa.

(2) The recovery could be practically full.

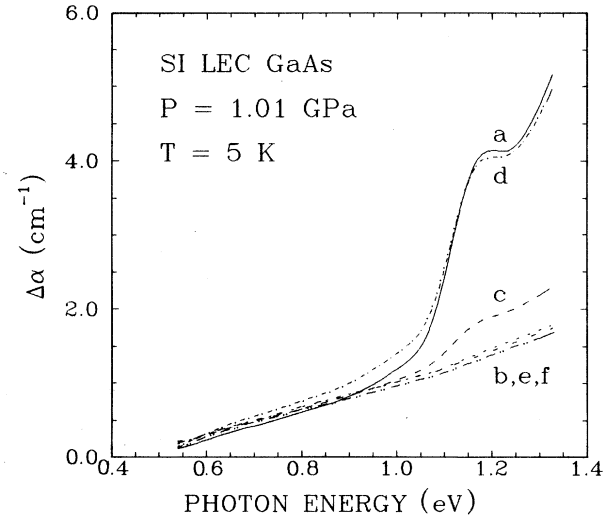

FIG. 2. The influence of the succeeding processes of the monochromatic light irradiation on the *EL2* absorption spectrm at 1.01 GPa for SI LEC GaAs at 5 K: (a) initial spectrum, (b) after 1.13-eV irradiation, (c) after 1.0-eV irradiation, (d) after 1.36-eV irradiation, (e) the same as (b), and (f) after 0.9-eV irradiation.

(3) The effect was a purely optical one because at 5 K all thermal recovery processes can be neglected.

(4) PIOR was reversible, i.e., photoquenching and PIOR processes could be repeated many times one after the other (see, e.g., Figs. 1 and 2).

(5) PIOR was effective for photon energies above as well as below (1.0 eV) the intra-$EL2^0$ absorption band, but it was not observed for $h\nu = 0.9$ eV (see Figs. 1 and 2).

(6) PIOR was very efficient—the time constant of the recovery being comparable with that of the photoquenching process. For example, for the *n*-type sample at $p = 0.36$ GPa the measured time constants were 60 s for the 1.13-eV photoquenching process, 80 s for the 1.36 eV, and 420 s for the 1.0 eV optical recovery processes, with the relative light intensities given by 1, 0.2, and 1.5, respectively. For the SI GaAs crystal PIOR was less efficient in comparison with the photoquenching process which enabled practically full photoquenching (see Fig. 2) in spite of the coexistence of both effects in the same photon energy range.

PIOR had qualitatively the same properties in both *n*-type and SI GaAs; however, simultaneously with the appearance of PIOR in the photoquenched spectra of the *n*-type crystal there was an additional absorption in the whole energy range investigated that was not present in the SI sample (see Figs. 1 and 2). It is clear from Fig. 1 that while the characteristic absorption of $EL2^0$ was photoquenched, the additional absorption increased, which suggests that this absorption is related to *EL2* in its metastable state. To verify this hypothesis we plotted the relative absorption coefficient $\Delta\alpha(1.2\text{ eV})$ vs $\Delta\alpha(0.85\text{ eV})$ taken from the absorption spectra measured for the *n*-type sample at $p = 0.36$ GPa [see Fig. 3 (some of these spectra are presented in Fig. 1)]. The linear correlation strongly supports our hypothesis. So, at high pressure the optical activity of the *EL2* defect in its metastable state manifests itself not only in the PIOR effect but also in the absorption related to the *EL2* defect in its metastable con-

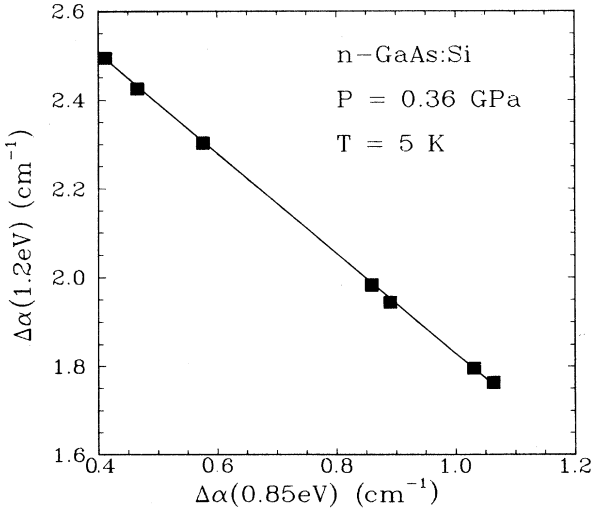

FIG. 3. The relative absorption coefficient $\Delta\alpha(1.2\text{ eV})$ vs $\Delta\alpha(0.85\text{ eV})$ taken from the absorption spectra measured for the *n*-type sample at $p = 0.36$ GPa.

figuration. Because the onset of PIOR at pressures $p > 0.3$ GPa was detectable neither in the pressure dependence of the intra-$EL2^0$ absorption[21] nor in the pressure dependence of the probability of the $EL2$ thermal recovery in SI GaAs (Ref. 22), one has to exclude any drastic rearrangement of the defect leading to the optical activity of its metastable configuration. The absorption related to the $EL2$ metastable state was observed only in the n-type crystal, not in the SI one. It extended over a wide energy range and its shape resembled a photoionization spectrum. We suggest that this absorption is the photoionization of the $EL2$ defect, which is its metastable configuration with an additional electron trapped on it. This means that at $p \geq 0.3$ GPa the $0/-$ level of the metastable $EL2$ enters the energy gap which, especially in n-type samples, enables the capture of an extra electron by $EL2$, resulting in the negative charge of the defect. This charge state of the metastable $EL2$ defect should be electron paramagnetic resonance (EPR) active and thus EPR studies could resolve the crucial problem of the microscopic structure of the metastable $EL2$ configuration. The capture of additional electrons by $EL2$ defects could be easily detected by Hall effect measurements. Such investigations are now in progress.

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