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Infrared absorption by deep levels in low-temperature electron-irradiated GaAs

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Semi-insulating GaAs was irradiated with 3-MeV electrons at 4.5 K and the optical absorption was monitored after irradiation and after annealing up to a temperature of 770 K. The absorption peak at 0.98 eV and the shoulder at 0.8 eV, which are known from room-temperature irradiation experiments, were observed immediately after irradiation at 4.5 K. Hence, the defect connected with these lines is produced directly during irradiation. The well-known recombination stages of Frenkel pairs located around room temperature and at 500 K lead to a reduction of the absorption background but do not affect the intensity of the two lines. This remarkable stability of the defect responsible for the absorption lines is further investigated by high-temperature irradiations and the final annealing between 600 and 650 K is attributed to reactions with mobile Ga vacancies.

The infrared absorption spectra of undoped semi-insulating GaAs are characterized by the dominant absorption line of the neutral change state of $EL2$ at 1.18 eV and an increasing structureless absorption close to the band edge. As observed by several authors,[1-6] the $EL2$ lines disappear after electron irradiation and defect lines, a peak at 0.98 eV ($P1$) and a shoulder at 0.8 eV ($P2$), appear in addition to a smooth background that increases strongly close to the band edge. These lines and their corresponding deep levels are the fingerprint of an irradiation induced defect that shows some very characteristic features. This defect seems independent of the doping of the GaAs (Refs. 1 and 2) (however, GaAs generally tends to become semi-insulating after high-dose irradiation) and of the dislocation density.[1] In contrast to this, the lines have not been observed after neutron irradiation[6] or after plastic deformation.[4,5] The defect seems to be stable up to about 600 K and the annealing kinetics of the lines has been investigated in detail; however, there is no clear correlation to other irradiation induced defects, e.g., the electron traps $E1$–$E5$ or the $EL6$ defect.[6] From the strong temperature dependence of the absorption line a large electron-phonon coupling was deduced and it was speculated that this defect should be a complex.[6]

All the irradiations have been performed at room temperature, i.e., above the temperature of a major annealing stage of irradiated GaAs that is attributed to the recombination of Frenkel pairs on the Ga sublattice.[7] In order to decide whether this defect complex is a direct irradiation product or a product of defect reactions that are due to the thermally activated defect migration—with the Ga interstitial atom most probably as the mobile species[8] —we initiated our low-temperature e^- irradiations.

Samples of undoped semi-insulating GaAs (thickness $d=0.39$ mm) were irradiated with 3-MeV electrons up to a dose of 3.5×10^{18} cm^{-2} at 4.5 K. They were transferred to the optical cryostat without intermediate warm-up. The optical absorption was followed during an isochronal annealing program up to 770 K. Measurements were done at a temperature of 2 K with photon energies from 0.4 eV to the absorption edge. The absorption coefficient α was computed from the transmitted intensities with and without the sample. Measurements of the magnetic circular dichroism (MCD) of the absorption were done in parallel[8] and will be reported in detail elsewhere; however, there seems to be no direct correlation between $P1$, $P2$, and a characteristic MCD signal.

Figure 1(a) shows absorption spectra as observed after irradiation and some steps of an isochronal annealing program with a holding time of 15 min at the different annealing temperatures T_a. The spectra clearly show the peak at 0.98 eV and the shoulder at approximately 0.8 eV superimposed on a featureless background. This background starts rising at about 0.6 eV. This background increases with irradiation dose and seems to be similar to the background observed after other types of irradiation. However, in contrast to high-dose neutron irradiations[9] our data cannot be well described by single exponential increase. The background and the peaks exhibit a distinctly different annealing behavior. As a measure of the background the absorption at 0.72 and 1.15 eV (i.e., outside the range of the peaks) is shown in Fig. 2(a) in the course of the isochronal annealing. At the lower energy, annealing already starts between 55 and 110 K; a stage around room temperature is found in both curves. In contrast, almost no change is seen around 500 K in Fig. 2(a); as shown in Fig. 1 there is, however, appreciable annealing at higher energies resulting in a shift of the absorption edge. The annealing stage around room temperature is usually ascribed to gallium Frenkel pairs, and the 500-K stage to arsenic Frenkel pairs.[8] Our results suggest that the contribution of the arsenic Frenkel pairs to the featureless background is mainly confined to higher photon energies (>1.3 eV), whereas gallium Frenkel pairs cause appreciable absorption at intermediate energies (around 1 eV) as well.

The same measurements were repeated after optical quenching of the samples at $T=2$ K with broadband light. In agreement with previous investigations[3,4,6] there are only minor changes observed, hence these defects do not show the metastabilities that are characteristic for $EL2$ and possibly some other As_{Ga} related defects.

We now turn to the absorption lines $P1$ and $P2$ that are superimposed on the smooth background. Two remark-

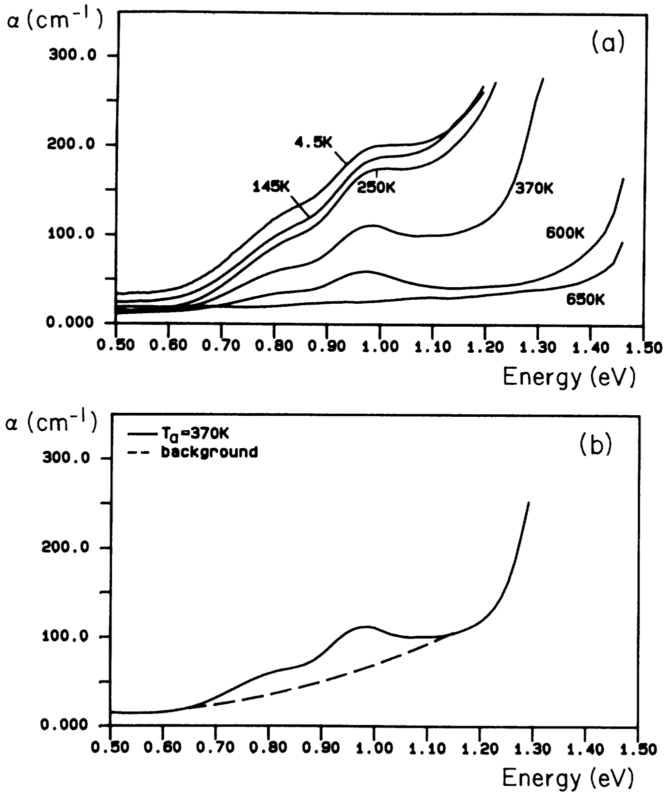

FIG. 1. Absorption coefficient of s.i. GaAs after electron irradiation at 4.5 K to a dose of $\phi t = 3.5\times10^{18}\ e^-/\mathrm{cm}^2$. The measurements were performed at 2 K and are shown after several steps of an isochronal annealing program. The lines at 0.98 and 0.8 eV are superimposed on a background that increases towards the band edge. (a) This background anneals continuously such that the range of energies that can be accessed by the experiment increases with the annealing temperature. (b) For the example of the annealing temperature of 370 K the extrapolated background line indicated was used for the evaluation of the height of the absorption lines.

able properties of the defects causing these lines are found here for the first time: (i) They are produced already at 4.5 K [see Fig. 1(a)] and (ii) they are stable all through the large annealing stage between 200 and 330 K. This stability is seen in Fig. 2(b), where the amplitude of the absorption line at 0.98 eV is normalized to the value after irradiation and plotted for the whole temperature range up to the final annealing at 650 K. The shoulder at 0.8 eV shows identical behavior. As the exact shape of the line and the background was not known, the background was assumed to be a smooth curve in order to separate the line. An example for the evaluation is shown in Fig. 1(b) for the 370-K curve. This uncertainty introduces errors not only to the absolute value of the absorption coefficient α but also to the annealing behavior because the background obviously anneals differently from the lines. Therefore, depending on the background subtractions, different annealing curves are published for α close to $E = 0.98$ eV.[1,2,6] Our procedures and results for the annealing of the line are very similar to those in Ref. 6, which were also obtained for semi-insulating GaAs.

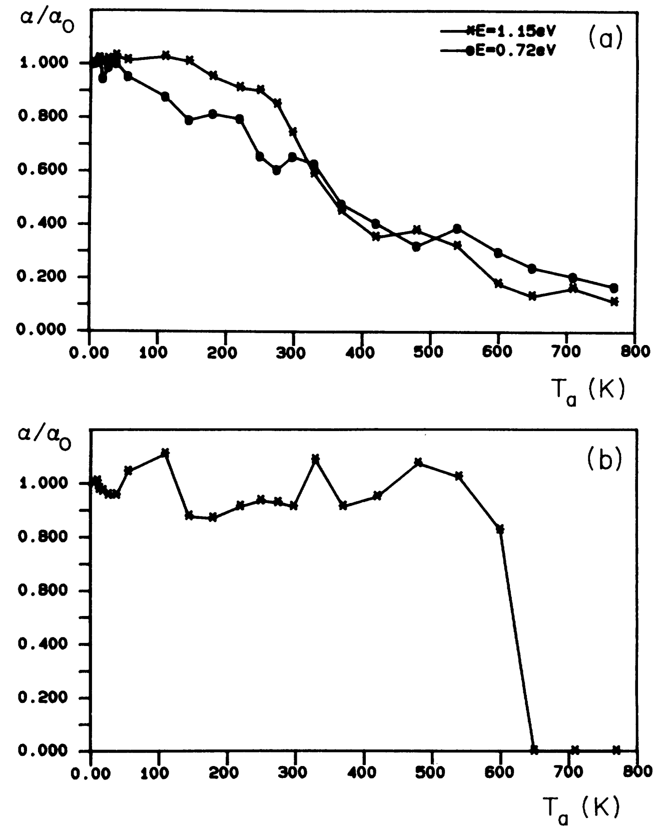

FIG. 2. (a) Annealing behavior of the background at low energies (0.72 eV) and high energies (1.15 eV) as compared to (b) the annealing of the absorption line at 0.98 eV. Values are normalized to the value α_0 observed directly after irradiation.

There is, however, some indication that the lines start to anneal at lower temperatures for p-type GaAs.[3]

In order to investigate this remarkable stability of the lines in further detail, we did some irradiations at higher temperatures. Figure 3 shows that after irradiation at 470 K, i.e., already within the 500-K stage, the defects are also formed. Considering the different irradiation dose the in-

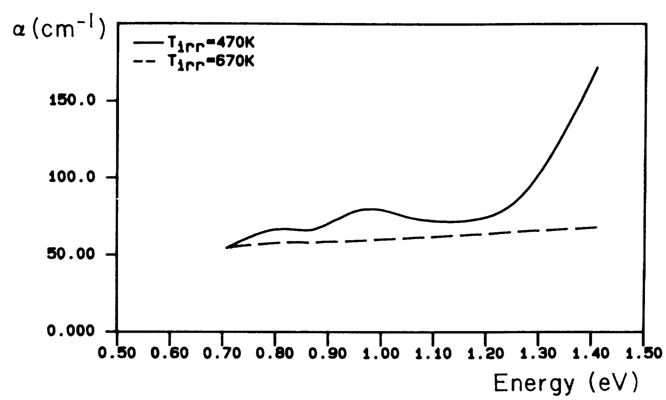

FIG. 3. Absorption coefficient (similar to Fig. 1) after electron irradiations at 470 K ($\phi t = 7.2\times10^{18}\ e^-/\mathrm{cm}^2$) and 670 K (normalized to the same dose).

troduction rate for the defects is reduced to about $\frac{1}{3}$ of the low-temperature values, but this reduction seems small compared to the reduction observed for other defect complexes [that is about $\frac{1}{10}$ (Ref. 10)]. This observation shows again that the defect must be produced directly by the irradiation particles and is not much affected by defect reactions that proceed thermally activated or triggered by the irradiation induced ionization events. As expected, the lines are no longer formed for irradiation temperatures above 650 K (Fig. 3).

These results impose some limits on structural models for the defect under consideration as well as on the annealing reactions. On the one hand, the low production temperature seems to exclude a larger cluster formed by agglomeration of mobile defects. On the other hand, it is very unlikely that an elementary point defect like a vacancy or an interstitial atom "survives" both large annealing stages, where mobile defects on both sublattices have been present and most of the radiation damage anneals; e.g., about 50% of the damage in the lattice parameter has annealed at 330 K and more than 80% at 550 K.[8] We suggest that the defect is a small complex immediately produced by irradiation.

A simple reason preventing such a complex from modification by mobile defects would be Coulomb repulsion of defects with the same charge state. Such a model is very similar to the generally accepted model for the close Frenkel pairs on the arsenic sublattice: As_i-V_{As};[6] these pairs cannot recombine up to a temperature of about 500 K although the free As_i is supposed to be mobile at much lower temperatures. Hence the complex discussed here needs only a slightly higher barrier in order to survive up to 600 K. Theory shows that three of the possibly mobile defects that would have been repelled are donors, i.e., Ga_i, As_i, and V_{As} and only V_{Ga} is an acceptor.[11-15] Since at least two of them have been repelled below 600 K (most probably Ga_i around room temperature and As_i and/or V_{As} at 500 K) we conclude that the complex must be a positively charged donor as well. Hence only the negative or neutral V_{Ga} is left as a defect that becomes mobile between 600 and 650 K and can eliminate $P1$ and $P2$. Although there is not yet a general agreement on the migration temperature of V_{Ga} our conclusion is in good agreement with the observation of the annealing of a signal that is attributed to V_{Ga} by investigation of the electron paramagnetic resonance.[16,17] The alternative possibility, which is that the complex dissociates at temperatures above 600 K, seems unlikely considering the observation that the annealing kinetics of $P1$ and $P2$ is characterized by a rather low activation energy and a large number of jumps of the mobile defect.[1,6]

The nature of the complex itself is more speculative at present. The complex Ga_{As}-As_i proposed in Ref. 6 seems unlikely as we would expect that the number of this type of complexes would be increased during the annealing stage at $T \approx 500$ K where the As_i is mobile. Considering that $P1$ and $P2$ are observed especially after electron irradiation we rather suppose that the complex could be the result of a short replacement collision chain as, e.g., V_{As}-As_{Ga}-Ga_i or V_{Ga}-Ga_{As}-As_i. Such a complex might be stabilized under the special ionization conditions during the collision and might not be formed anymore during thermal annealing. After the arrival of an additional V_{Ga}, this kind of complex reduces to a single vacancy V_{Ga} that is mobile and can undergo further reactions. However, more experimental as well as theoretical work seem necessary for a detailed discussion.

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