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E.R. Weber and P. Omling

April 1985

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Prepared for the U.S. Department of Energy
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ANTISITE DEFECTS AND EL2 IN GaAs

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Abstract

The characteristic properties of anion antisite defects and the technologically important midgap donor EL2 in GaAs are reviewed with emphasis on information concerning the microscopic structure of these defects. The wide body of existing results points towards As_{Ga} as being involved in the EL2 defect. However, there is evidence that both antisite defects as well as EL2 are members of defect families with slightly varying properties and microstructure. At present it is not yet possible to determine the detailed structure of EL2 and the other midgap donor levels in GaAs.

Introduction

The most important and most investigated defect in GaAs is the midgap donor level EL2, which is responsible for the compensation of bulk GaAs grown in Si-free crucibles. Therefore it is of technological as well as scientific interest to identify the microstructure of this defect and to determine its characteristic properties. For the last few years, anion antisite defects have been discussed as possible defects producing the EL2 level. In this paper, our present knowledge of the properties of anion antisite defects as well as of EL2 will be reviewed, including most recent experimental results. Based on these data, the possible conclusions as to the microscopic nature of these defects will be discussed.

Antisite Defects in GaAs

Anion antisite defects are the first intrinsic lattice defects identified by EPR in GaAs[1-5]. They are present as native defects in as-grown material[1] and their concentration can be enhanced by electron[2] or neutron irradiation[3,5], or by plastic deformation[4]. Figure 1 shows EPR spectra of the characteristic quadruplet observed in as-grown, plastically deformed, and in n-irradiated semi-insulating LEC GaAs. These spectra show no measurable anisotropy. They can be satisfactorily described by a S=1/2 center with a I=3/2 nuclear spin of 100% abundance in an A_1 state. The observed nuclear spin together with the defect production by irradiation (up to $10^{18}cm^{-3}$) excludes the possibility of impurities and leaves only an As-related defect as origin of this signal.

Due to the tetrahedral symmetry the possible defect models are limited to an As interstitial (As_i) or the As_{Ga} antisite. The first possibility can be rejected based on the following arguments: (1) As_i, like P_i in GaP, is expected to have a ground state of T_2 symmetry[6], whereas for As_{Ga} an A_1 ground state has been predicted[7-9], in agreement with experiment. (2) The observed hyperfine splitting of 0.0905 cm^{-1} is 0.94 of the hyperfine splitting of P_{Ga} in GaP (0.0966 cm^{-1}). For comparison, the ratio of the isotropic hyperfine interaction for the AsO_4^{4-}/PO_4^{4-} radicals is 0.95, and a simple theoretical consideration, using neutral-ion Hartree-Fock wavefunctions, gives 0.94 as well[1]. (3) This defect shows high thermal stability[10,11] up to 950°C, in contrast to the usually observed annealing of interstitial defects in semiconductors at or below room temperature.

The As_{Ga}^{4+} spectrum consists of lines of width 250-350G. This large line

width is most likely due to the unresolved superhyperfine-interaction with the four neighboring As atoms, splitting each of the four quadruplet lines into 13 components. In the case of such broad lines it is not possible to distinguish from EPR measurements alone, if the defect has perfect tetrahedral symmetry or if it is weakly distorted, e.g. by another defect on the next-nearest shell, like V_{Ga}.

In addition to the As_{Ga}^{4+} quadruplet, a singlet line near g=2 appears in the spectra of As_{Ga} in n-irradiated GaAs[3,4,5], see Fig. 1. This line has been tentatively ascribed to V_{Ga}^{2-} [5]. Its occurrence together with the As_{Ga}^{4+} quartet has been taken as evidence that both defects might indeed be coupled to one complex, producing both spectra. However, recent experiments[12] demonstrated independent annealing behavior for both spectra, so that they might well arise from independent defects produced together during e.g. n-irradiation. The spectra shown in Fig. 1 allow a further observation: The relative intensity of the singlet and the quadruplet depends on the defect concentration. In as-grown material the third line of the quadruplet amounts in height to 1.3 of the neighboring lines, in strongly n-irradiated material it increases up to 2.6 of its neighbors'. Thus the defect responsible for the singlet is produced together with As_{Ga}, however not with the same introduction rate.

In addition to the singlet described in Ref. 5, as-grown GaAs exhibits a further broad line near g=2 with a noticable, unresolved anisotropy, called GaAs-BE1, see Fig.2. Up to now, this spectrum has not yet been described. It appears only after photoquenching of the As_{Ga}^{4+} signal in as-grown GaAs. Experiments are currently in progress to determine, whether the GaAs-BE1 spectrum is due to an excited triplet state of As_{Ga}^{4+} (the D° state) or to

another defect which becomes paramagnetic by the change of the Fermi level during illumination.

Anion antisite defects in III-V semiconductors, group V atoms on group III sites, can be expected to be double donors. The large hyperfine splittings of the antisite defects, compared e.g. with the shallow, delocalized donors in Si, indicate directly the localized nature of these defects. Thus they can be expected to be deep level defects. Photo-EPR results of As_{Ga}^{4+} in plastically deformed, semi-insulating LEC and p-GaAs are shown in Fig. 3[4,13]. The two quenching steps at 0.75eV and 1.05eV have been interpreted in terms of a double donor model for As_{Ga}: The concentration of paramagnetic As_{Ga}^{4+} (D^+) is decreased by filling the single donor level with 0.75eV light. This quenching is not complete, because 0.75eV illumination allows as well emptying of the level to the conduction band. At 1.0 eV, a further decrease of As_{Ga}^{4+} can be noticed, because then, in addition, emptying of the double donor level to the conduction band is possible. This model gains support from the photo-response of p-GaAs, which exhibits an enhancement step near 0.5eV, that can be attributed to the filling of the partially occupied double donor level of As_{Ga}. This level scheme recently found additional support by photo-capacitance measurements, giving direct evidence for the existence of the double donor level near E_v+0.5eV[14]. Self-consistent pseudopotential[7] as well as tight binding calculations[8] were in agreement with this assignment.

The results of thermal annealing of the As_{Ga}^{4+} spectrum in as-grown, n-irradiated and plastically deformed GaAs have been discussed recently[10]. Whereas As_{Ga}^{4+} is thermally stable up to 950°C in as-grown material, it anneals partly in plastically deformed or n-irradiated samples at 450°-600°C.

This behavior points to a complex nature for this defect, see below.

The midgap donor level observed for As_{Ga} is at the same energy as the technologically very important midgap donor "EL2"[15] in GaAs. Before further discussion of the microscopic nature of the As_{Ga}^{4+} spectrum, we will briefly review the relevant properties of this defect and discuss the possible relation of EL2 to As_{Ga} antisite defects in Section 4.

EL2 in GaAs

The mid-gap deep donor level EL2 is the dominant electron trap in both bulk grown and VPE grown GaAs material. The concentration of this defect is determined by the stoichiometry conditions during growth, with high concentrations under As-rich conditions[16-18]. This indicates the presence of native defects in the formation mechanism. The importance of the EL2-level from the device point of view, is the compensating role it plays in undoped liquid encapsulated Czochralski (LEC) and Bridgman grown bulk material, leading to the formation of semi-insulating (SI) GaAs[19]. Such material is promising for further development of fast GaAs integrated circuits.

Besides this technical aspect, the EL2-level shows very exciting physical properties. For instance, when a photocapacitance experiment is carried out (using photons in the range $0.75<h\nu<E_g$) at temperatures higher than T=150 K (but lower than T=250 K to avoid thermal emission processes) on EL2 levels filled with electrons, the capacitance signal increases monotonically to a final value, corresponding to curve (a) in Fig. 4. Performing the same experiment at low temperatures (T<100 K) the signal increases, but instead of reaching the final value observed at T>150 K the capacitance signal decreases

(with a time constant that is ~100 times larger than that of the increase of the signal) approaching the initial capacitance value (curve (b) in Fig. 4). This is the photocapacitance quenching effect of EL2 in GaAs[20]. It has been concluded that the effect is caused by the transformation of the "normal" EL2 center, $EL2^0$, into the metastable state, $EL2^*$, of the same charge state. Once transformed to the $EL2^*$ state, the center is insensitive to further illumination. The spectral distribution of the optical transition rate for the $EL2^0 \rightarrow EL2^*$ transition is shown in the insert of Fig. 4. The reverse transition ($EL2^* \rightarrow EL2^0$) can only be performed by heating the sample to T>140 K or injecting carriers at T>30 K. Both processes are temperature activated and the regeneration rate can be described by[21,22]:

$$r = 3.6\ 10^{12}\exp(-0.36[eV]/kT) + 2.8\ 10^{-14}n\langle v_{th}\rangle\exp(-0.106[eV]/kT)\ [s^{-1}] \quad (1)$$

where $\langle v_{th}\rangle$ is the thermal velocity of the free electrons and n the free electron concentration.

The metastable properties of the EL2 level were originally explained by Bois and Vincent[23] as a strong lattice relaxation effect which could be visualized in a configurational coordinate diagram. Recently Levinson[24] proposed a new model which is based on a charge interaction between one shallow and one deep donor within a complex. This model explains most of the observed electronic properties of the EL2 level, and has been strongly supported by thermally stimulated current measurements performed by Fillard et al.[25].

The identification of the EL2 level is often performed by thermal emission rate measurements, absorption measurements or photocapacitance quenching

experiments (or similar EL2-bleaching experiments). It has recently been questioned as to whether these identification methods are unambiguous. For instance, in early studies[26] it was observed that different GaAs samples showed different thermal emission rates for the EL2 level. Later Watanabe et al.[27] observed that different growth conditions during MOVPE growth of GaAs created two different EL2 levels, $EL2_a$ and $EL2_b$. Recently these two EL2-levels were also observed in Bridgman grown GaAs by Lagowski et al.[28], who also found a dependence on the formation of one of the levels, ELO (=$EL2_b$), on the amount of oxygen added during growth. The oxygen correlation might, however, be an indirect effect. These results are presented in Fig. 5. The thermal emission rate[28,29]:

$$e^{-1} T^2 = 3.53 \cdot 10^{-8} \exp (0.815 \text{ [eV]}/kT) \qquad [s^{-1}] \qquad (2)$$

is probably the most accurate fingerprint of the EL2 level in conducting material. Unfortunately, this fingerprint cannot be used easily for identification of EL2 in semi-insulating material, where the best method to identify and determine concentrations are absorption measurements.

The relation between the absorption coefficient for certain wavelengths and the EL2 concentration (as determined by electrical methods) was established by Martin[30]. This technique is widely used but has some problems associated with it. The first is the fact that absorption measurements are not selective measurements, i.e. the absorption coefficient is the sum over all possible absorption processes in the material. This might lead to overestimates of the EL2 concentration, and to distortion of the spectral shape of the absorption spectrum. Recently, Samuelson et al.[31] presented a method which allowed selective measurement of the EL2 absorption,

thereby making the EL2 analysis more reliable. The second problem is the Fermi level position. As pointed out by both Martin[30] and Walukiewicz et al.[32] the shape of the absorption spectra are strongly dependent on whether the Fermi level is above or below the EL2 levels. The ideal situation is to determine the Fermi level position by use of Hall effect measurements. It is, however, always safe to use "n-type" material.

The observation of the quenching effect has for a long time been one of the strongest argument for an EL2 identification. It has, for instance, been used to identify several luminescence properties as being EL2 related. During recent years there has been an increasing concern over the possibility that the observed quenching in these experiments are only indirect effects. The following two cases are examples, where the observed quenching properties are caused by two different indirect effects.

The first case is the 0.635 eV luminescence band in GaAs which was suggested to be EL2 related due to its quenching properties under extrinsic excitation[33]. However, Omling et al.[34] showed that the extrinsic excitation of the luminescence was performed via the EL2 level, and therefore quenching of the EL2 level caused an apparent quenching of the 0.635 eV luminescence.

The second case is the observation that the paramagnetic charge state (Fe^{3+}) of the substitutional Fe_{Ga} impurity can be detected, only if the EL2 levels in SI as-grown GaAs are quenched[35]. Since it is well established that the ground state of the non-paramagnetic Fe^{2+} level is located at E_v+0.5 eV[36], it is concluded that the Fermi level has dropped at least below this level. In fact, one can measure the quenching properties of the

EL2 level on the Fe^{3+} EPR spectrum, and with great confidence conclude that the effect is indirect.

Finally it should be mentioned that photocapacitance measurements of the photoionization cross section σ_n can also be used for identification of the EL2 level. The σ_n spectrum of EL2 shows very characteristic structures which have been interpreted as a mixture of deep level to different conduction band transitions[37] and internal transitions[38]. This method has been used by Taniguchi et al.[39] to show that there exist many different EL2 levels in different materials. It should, however, be noted that this method is very sensitive to the kind of substrate used for measurements on epitaxial layers (e.g. self absorption), and to other deep levels present in the crystal. The last point is particularly true in bulk samples where several defects are always present.

The Microscopic Identification of EL2 in GaAs

The discussion in the preceding paragraphs mentioned already some of the arguments suggesting a correlation of EL2 with As_{Ga} antisite defects in GaAs: The stoichiometry dependence of EL2 formation is consistent with the As_{Ga} model[16-18]. Antisite defects are found in as-grown material in concentrations comparable to EL2. Photo-EPR found clear evidence for a midgap donor level of As_{Ga} [4]. The concentration of As_{Ga} as well as EL2 has been reported to increase in plastically deformed[4,40,41] as well as in n-irradiated[3,10,42-44] GaAs. The thermal stability of EL2 and As_{Ga} in as-grown GaAs up to 950°C is well documented[10,11,45]. The persistent photoquenching properties of EL2 have been observed for the As_{Ga}^{4+} EPR signal in plastically deformed GaAs as well[4,46]. Figure 6 shows thermal de-quenching rates for EL2[21,22] and

As_{Ga} [46] to be consistent. Altogether, these arguments give strong evidence for a correlation between EL2 and As_{Ga} defects. They are, however, no final proof that EL2 is indeed a donor level of ideal, undistorted antisite defects.

Recent absorption measurements performed by Kaminska et al.[38] revealed a fine structure with a zero-phonon line at 1.04 eV. This structure was claimed to be a strong evidence for the identification of EL2 as being the neutral charge state of an undistorted antisite defect. The identification of the lines as being internal antisite transitions are probably correct, but it was recently shown by Lagowski and Gatos[29] that the intensity of the zero-phonon line was not correlated to the EL2 concentration, which seems to be inconsistent with the direct EL2 -As_{Ga} identification. This fine structure has also been observed in very recent photo-current experiments[47], in contradiction to what was originally claimed[38].

There are various further arguments against an identification of EL2 with ideal, undistorted As_{Ga} centers[29,48]. First, it is difficult to explain the persistent photoquenching effects without taking into account additional defects leading to the proposed large lattice relaxations in the quenched state. A model proposed by Baranowski et al.[49] of possible relaxations of undistorted As_{Ga} defects is obviously in direct contradiction to self-consistent calculations by Scheffler et al.[50]. Recent measurements of the magnetic circular dichroism (MCD) induced by As_{Ga} seem to contradict the EL2 - As_{Ga} identification[50]. Recent measurements of the absorption quenching in plastically deformed and n-irradiated GaAs (see Fig. 7) suggest as well different microscopic structures for EL2 and the antisite defects

introduced by plastic deformation or irradiation.

On the other hand, it has already been discussed above that the EPR spectrum alone does not allow one to conclude an ideal, isolated As_{Ga}^{4+} as origin of this spectrum. Due to the large line width, a complex of As_{Ga} with other defects might give rise to at least part of the spectra. A careful analysis of the temperature dependence of the As_{Ga}^{4+} spectrum[5,12] revealed evidence for more than one defect contributing to the EPR quadruplet spectrum of As_{Ga}. The different annealing behavior of As_{Ga}^{4+} in as-grown compared to plastically deformed or n-irradiated GaAs[10] indicates as well that more than one center can produce very similar EPR spectra. The recent optically detected ENDOR investigations[51,52] are the first direct proof for this notion. However, due to the lack of quantitative ENDOR spectra, it is not yet possible, to assess the fraction of undistorted As_{Ga} in the total spectrum. Such a family of antisite defects is already well known in GaP, where the smaller line width allows distinction of different antisite-complexes by EPR alone[53].

Finally, it appears to be appropriate to express a word of caution towards the quantitative interpretation of As_{Ga} EPR spectra, e.g.[54,55]. EPR detects only the paramagnetic charge state As_{Ga}^{4+} of As_{Ga} defects, thus the signal intensity can depend on the Fermi level in the sample at the measurement temperature. Appearance of an Fe^{3+} spectrum[54] is in any case an indication for a shift of the Fermi level down to E_v+0.5 eV, as discussed above. Therefore it is essential to include the possibility of Fermi level variations in the interpretation of quantitative EPR data.

Conclusion

The discussion of the properties of As_{Ga} defects and EL2 in GaAs have revealed many similarities, indicating a correlation of these defects. However, there is growing evidence that ideal, undistorted antisite defects are not responsible for EL2 with its persistent photoquenching behavior. Optically detected ENDOR investigations[51,52] demonstrate the existence of undistorted as well as distorted antisite defects, similarly to the well-known family of anion antisite defects in GaP. On the other hand, there is convincing evidence of the existence of more than one midgap donor level, so that the family of antisite defects might correspond to a family of midgap donor levels. Future investigations, including ENDOR measurements, will have to identify direct correlations between well characterized members of these defect groups.

Acknowledgments

This work was supported by the Director, Office of Basic Energy Research, Office of Basic Energy Sciences, Material Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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Figure Captions

Fig. 1. EPR spectra of As_{Ga}^{4+} in as-grown, plastically deformed and n-irradiated GaAs. The As_{Ga}^{4+} concentrations range from $5 \cdot 10^{15} cm^{-3}$ (as-grown) to $1 \cdot 10^{18} cm^{-3}$ ($3 \cdot 10^{17}$ n/cm^2). The spectra in as-grown and plastically deformed samples were taken with a computer for background subtraction.

Fig. 2. EPR spectrum of a new resonance (GaAs-BE1) with unresolved anisotropy in as-grown GaAs, appearing after photo-quenching of As_{Ga} by white light illumination. Superimposed on this line is a weak Fe^{3+} spectrum (narrow lines).

Fig. 3. Photo-EPR of As_{Ga}^{4+} in a) s.i. LEC GaAs and b) p-GaAs[4,13]. The insert shows the level scheme proposed for the As_{Ga} double donor[4].

Fig. 4. Photocapacitance transients observed with EL2 in GaAs: a) sample tem-

perature between 150 K and 250K, showing the fast transient, b) sample temperature lower than 100 K, showing in addition the slow transient from the photocapacitance quenching effect[20]. The insert contains the spectral distribution.

Fig. 5. Arrhenius plot of electron emission rates (DLTS) of "EL2" in MOVPE GaAs ($EL2_a$ and $EL2_b$,[27]) and in Bridgman grown GaAs (EL2, ELO,[28]).

Fig. 6. Thermal regeneration rate of quenched EL2 (Photocapacitance measurements, o, ● :[21], : Δ[22]) and of quenched As_{Ga}^{4+} (EPR, □ :[46]).

Fig. 7. Optical absorption spectrum of plastically deformed GaAs (2.5 % at 400°C), measured before (α) and after (α_Q) quenching of EL2.
The quenched fraction of the absorption shows the spectral dependence characteristic for EL2[34].

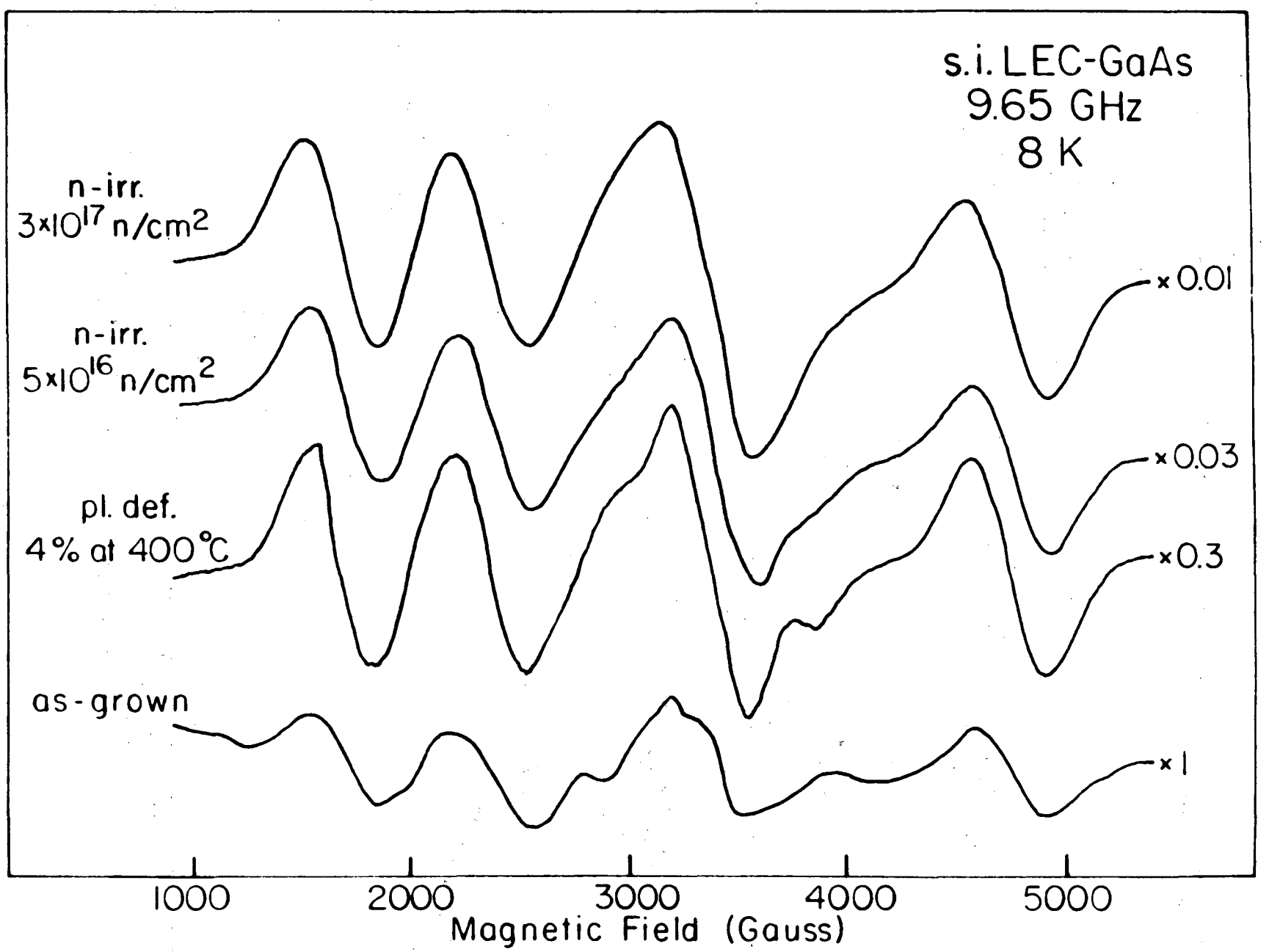
s.i. LEC-GaAs
9.65 GHz
8 K
n-irr.
3×10^17 n/cm^2
n-irr.
5×10^16 n/cm^2
pl. def.
4% at 400°C
as-grown
×0.01
×0.03
×0.3
×1
1000
2000
3000
4000
5000
Magnetic Field (Gauss)

Figure 1

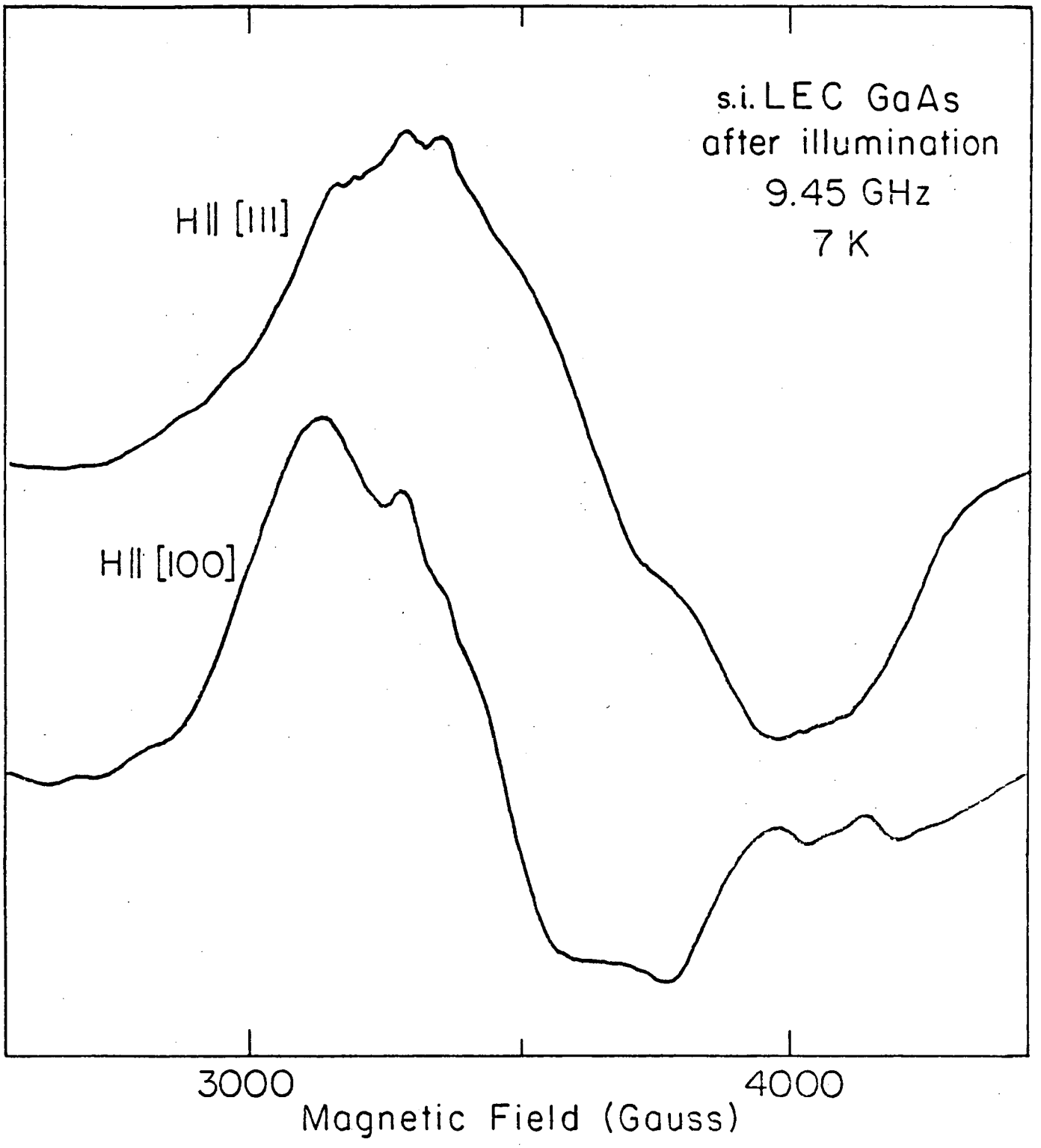
s.i. LEC GaAs
after illumination
9.45 GHz
7 K
H || [111]
H || [100]
3000
4000
Magnetic Field (Gauss)

Figure 2

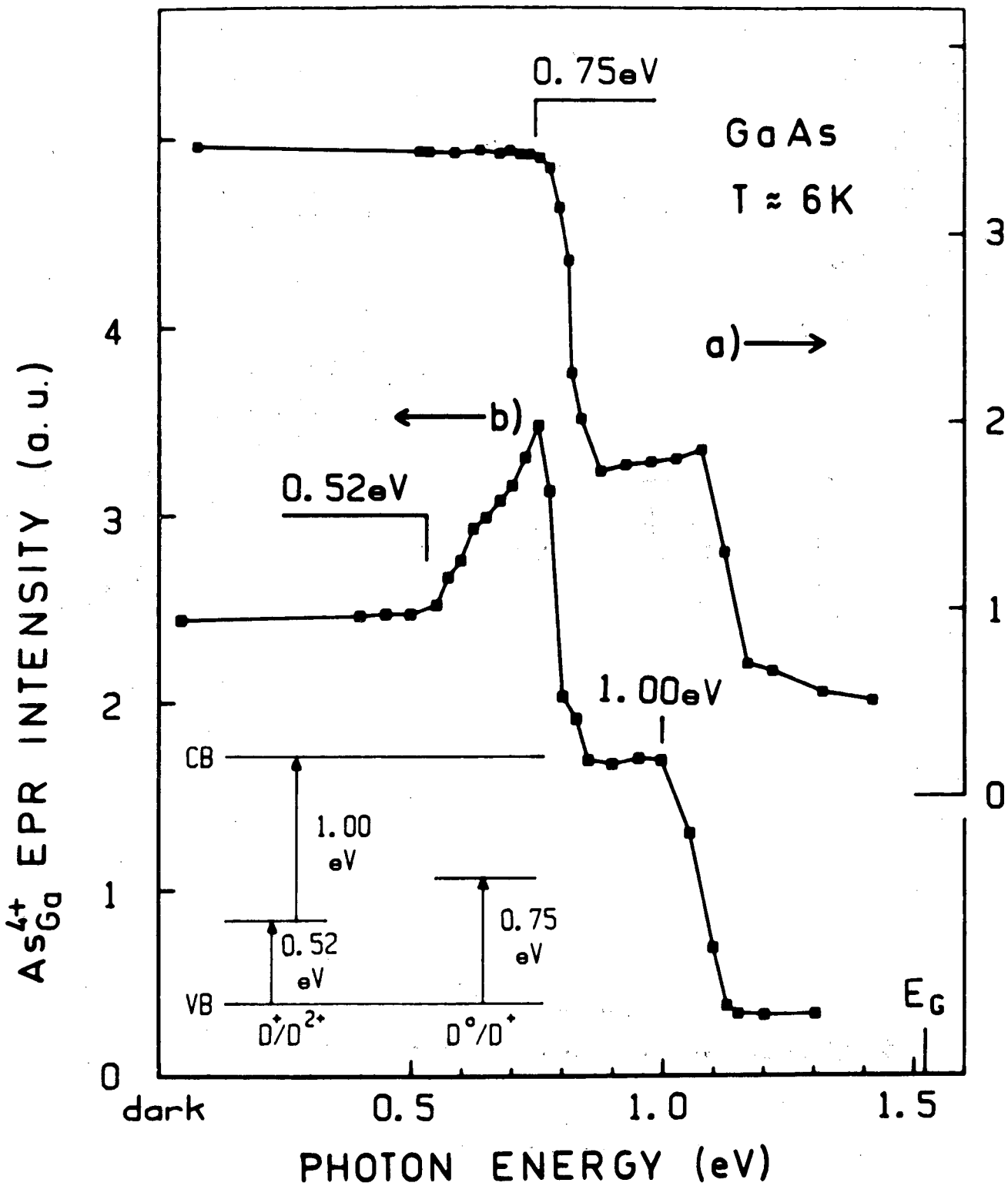
0.75eV
GaAs
T ≈ 6K
a)
b)
0.52eV
1.00eV
CB
1.00
eV
0.52
eV
VB
0.75
eV
D+/D2+
D°/D+
EG
As4+Ga EPR INTENSITY (a.u.)
PHOTON ENERGY (eV)
dark
0.5
1.0
1.5
0
1
2
3
4

Figure 3

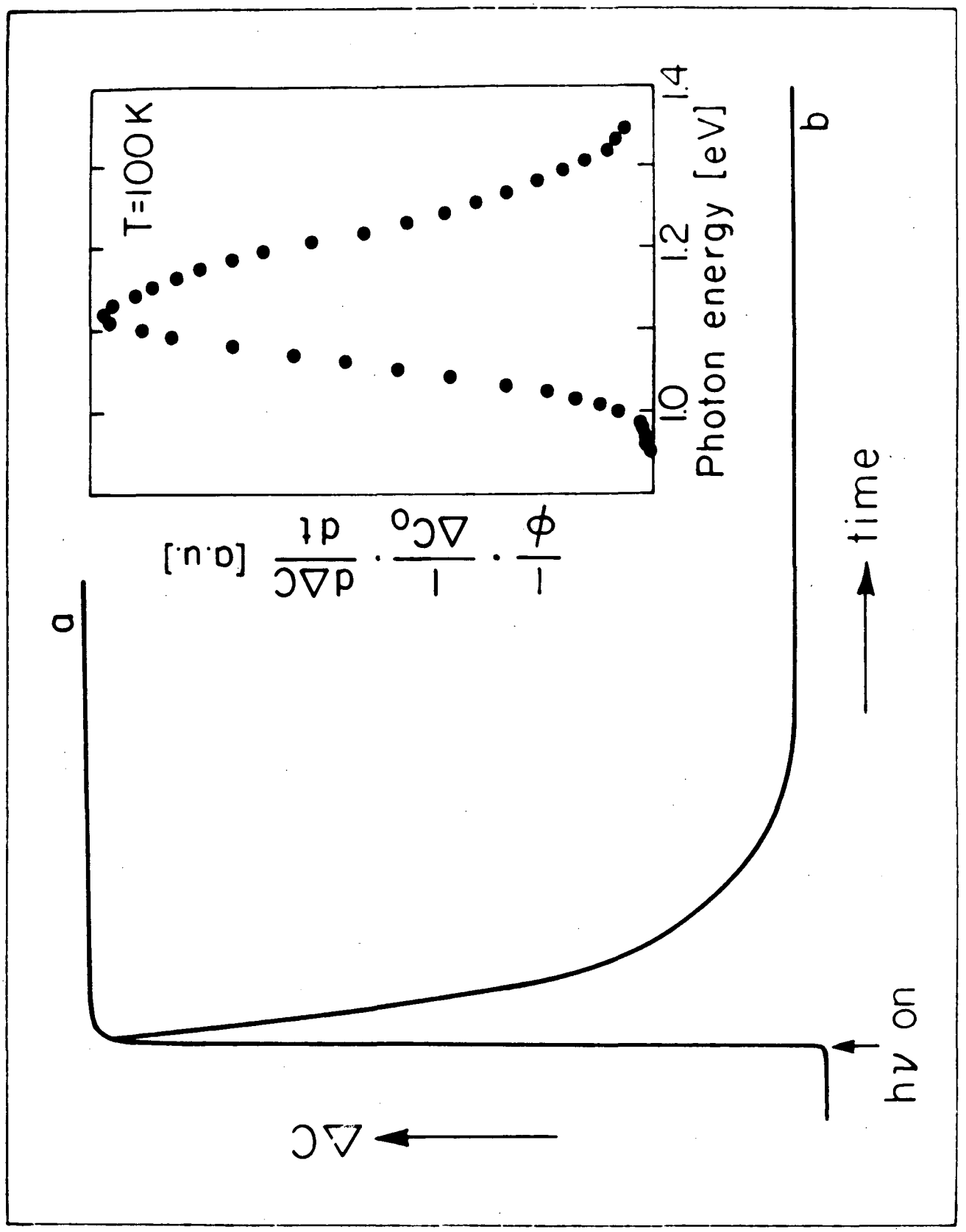
a
b
ΔC
hν on
time
T=100 K
1.0
1.2
1.4
Photon energy [eV]
$\frac{1}{\phi} \cdot \frac{1}{\Delta C_0} \cdot \frac{d\Delta C}{dt}$ [a.u.]

Figure 4

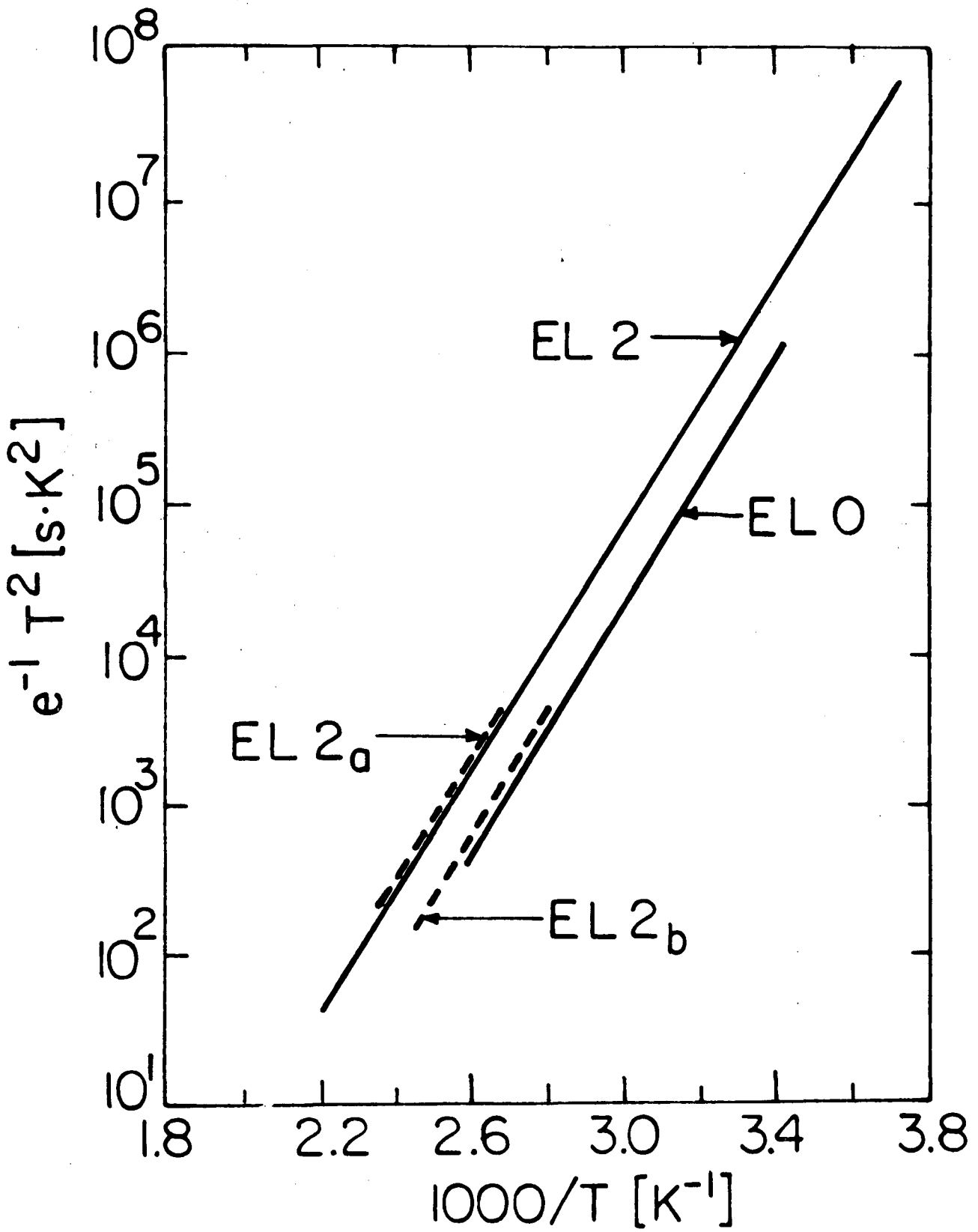
10^8
10^7
10^6
10^5
10^4
10^3
10^2
10^1
$e^{-1} T^2 [s \cdot K^2]$
EL 2
EL 0
EL 2_a
EL 2_b
1.8
2.2
2.6
3.0
3.4
3.8
$1000/T [K^{-1}]$

Figure 5

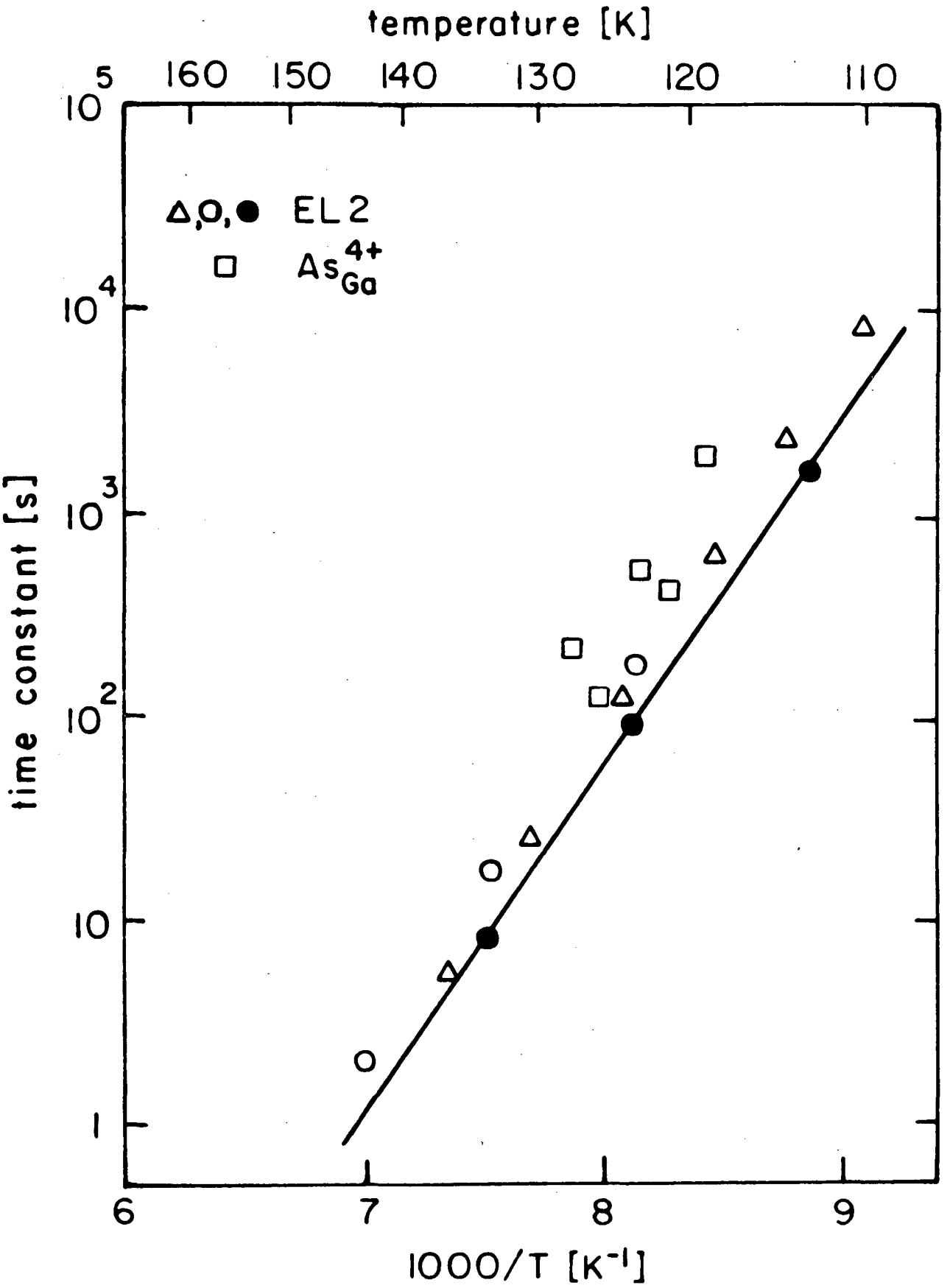
temperature [K]
160
150
140
130
120
110
10^5
10^4
10^3
10^2
10
1
time constant [s]
Δ,O,● EL 2
□ As_{Ga}^{4+}
6
7
8
9
1000/T [K^{-1}]

Figure 6

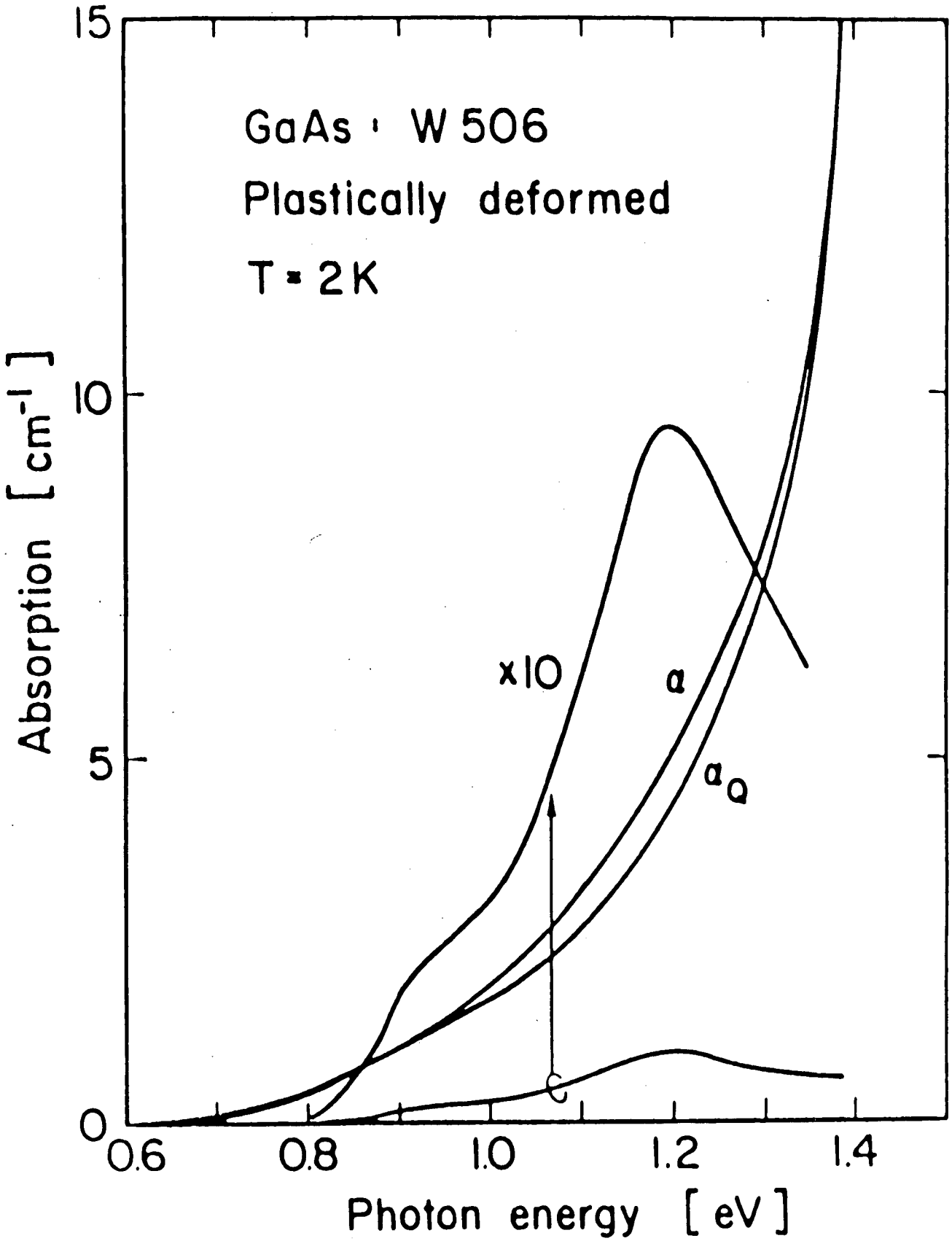
GaAs : W 506
Plastically deformed
T = 2K
x10
α
α_Q
Absorption [cm^{-1}]
Photon energy [eV]
0
5
10
15
0.6
0.8
1.0
1.2
1.4

Figure 7

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