Characterization of the EL2 center in GaAs by optical admittance spectroscopy

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We have measured the electron optical capture cross section, $\sigma_n^0(h\nu)$, of EL2 (the most important native center in GaAs) using a new technique which we have recently developed: optical admittance spectroscopy. This is a spectroscopic technique based on the measurement of the capacitance and conductance of a junction under monochromatic light of energy $h\nu$. This technique allows the measurement of the spectrum $\sigma_n^0(h\nu)$ of each center located in the band gap. We have measured the electron photoionization cross section of the EL2 center, $\sigma_n^0(h\nu)$, at three different temperatures within a range limited at high temperature by thermal emission and at low temperature by photoquenching (a feature characteristic of EL2 below 140 K). The study of the experimental data reveals that this center has a more complex nature than that of a simple defect. It seems to behave like a family of very close levels corresponding to similar atomic structures and located near the midgap. These results also reveal the existence of a shallow level close to the valence band and associated with EL2.

I. INTRODUCTION

EL2 is one of the most important native centers appearing in GaAs. It is located near the midgap ($E_C - 0.76$ eV), and it plays an important role in the growth of semi-insulating GaAs. It is also widely known for its peculiar behavior. This explains the considerable amount of work published about this center. It appears in GaAs, irrespective of the growth technique employed, except when it has been grown by liquid phase epitaxy (LPE) in a Ga-rich atmosphere or by molecular-beam epitaxy (MBE) (in this case probably due to the low growth temperature). EL2 can also be created by plastic deformation, by ionic implantation, or by neutron irradiation followed by an anneal at temperatures higher than 450 °C.

Although there have been many studies about EL2, its physical origin remains unknown. Initially, it was thought[1] that it was an oxygen-related center. This hypothesis was rejected by other authors.[2–4] The most commonly accepted theory at the moment relates EL2 with the arsenic antisite As_{Ga}.[5,6] Different experimental results as well as the complex phenomenological behavior of EL2 reveal a nature which does not correspond to that of a simple defect. It is more like a family of levels located near the midgap and related to the arsenic antisite. An example of this type of family is the resulting association of arsenic antisite with the arsenic vacancy V_{As}, the gallium vacancy V_{Ga}, or interstitial arsenic I_{As}, etc. These conclusions were drawn by Mochizuki and Ikoma[7] based on the nonexponentiallity of the transients of the optical recovery of photoquenching. These authors explain this family of levels by an arsenic cluster model.

Taniguchi and Ikoma[8] have revealed by deep-level transient spectroscopy (DLTS) a set of traps located in the midgap, which can be classified into two groups. Traps belonging to the first group are rather stable in their properties, whereas those belonging to the second group are not.

Other works[9,10] showed that oxygen creates a level different from EL2, but with about the same energy: the EL0. This center has a capture cross section 4 times that of EL2. These two centers cannot be distinguished by DLTS because of their proximity in energy.

The relation of EL2 with the arsenic antisite As_{Ga} is confirmed by the results obtained by electronic paramagnetic resonance (EPR). Theoretical considerations suggest that the As_{Ga} must appear as a double donor whose higher-energy level, located at the midgap, would be the fundamental level of EL2. Its lower level has been identified by Wosinski[11] and Lagowski *et al.*[12] at an energy of 0.45 eV above the valence band. Its donor nature has been confirmed by Mircea *et al.*[13] by DLTS and standard C-V profiling at high and low temperatures. Mittoneau *et al.*[14] have determined an activation energy for the EL2 center of $E_C - 0.825$ eV. These authors have also shown that the thermal capture cross section has an activation energy of 65 meV, which yields an ionization energy $E_T = 0.76$ eV for the EL2 center. The variation of the thermal capture cross section with temperature reveals the existence of strong lattice relaxation during the capture process.

From the phenomenological point of view, the most peculiar behavior of EL2 appears when studying its optical properties. Because of the midgap position of this center, transitions to both bands occur when the sample is illuminated. A more careful interpretation of the experimental data is then required to separate the contribution of each transition. The electron and hole optical capture cross sections σ_n^0 and σ_p^0 were first measured by Chantre, Vincent, and Bois[15] by deep-level optical spectroscopy (DLOS). These authors have developed a semiempirical model which matches their DLOS data acceptably well for photon energies below 1.4 eV. Above this energy their model predicts a decrease, in contradiction with their experimental results. Morante *et*

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al.[16] have measured the optical capture cross section of EL2 by optical isothermal transient spectroscopy (OITS), and their results agree with Chantre and co-workers' model even at energies above 1.4 eV. Another useful technique for the study of EL2 is the dual-source differential photocapacitance (DSDP) introduced by White *et al.*[17,18] This technique allows the selective measurement of both electron and hole optical capture cross sections. It has been found[19–21] that the electron optical capture cross section is always higher than the hole optical capture cross section at any photon energy $h\nu$. The most characteristic optical behavior of EL2 is the photoquenching, first observed by Bois and Vincent.[22] This phenomenon occurs when a GaAs sample, at a temperature lower than 100 K and containing the EL2 center, is illuminated with a monochromatic light of photon energy between 0.9 and 1.35 eV. A capacitance increase, corresponding to the normal photoionization transient of an initially filled center, is induced. However, the capacitance reaches a maximum and then decays with a time constant two orders of magnitude longer than the initial rising transient, reaching a final value close to that existing before illumination. After that, the center becomes insensitive to any further change in illumination. The optical recovery of the center cannot be achieved not even by leaving the sample in the dark. There are two mechanisms to regenerate the initial optical properties of EL2: thermal regeneration and electrical regeneration. In the first case the sample is held in the dark at temperatures between 100 and 130 K. The center gradually changes from its metastable state to the optically active state following an Arrhenius law with an activation energy of 350 meV. There is no photoquenching at temperatures above 140 K. At temperatures below 100 K, optical regeneration of EL2 can be achieved by a direct bias pulse which creates a high concentration of electrons in an n-type semiconductor. This process follows an Arrhenius law with an activation energy of 107 meV. The photoquenching efficiency varies with the photon energy within the range $0.9 \leqslant h\nu \leqslant 1.35$ eV, indicating a maximum which depends on the temperature and is located at about 1.1 eV. The height and position of this maximum decrease when temperature is increased. Another photoquenching phenomenon has also been observed[23] at energies above 1.35 eV when the sample has been previously illuminated with a light of energy $h\nu \approx 1.15$ eV. Nojima[24] also observed slow relaxation processes in photoconductivity measurements at an energy $h\nu = 0.8$ eV in undoped semi-insulating LEC GaAs which can be attributed to an optical transition from the metastable state to the conduction band via the fundamental state.

Several theoretical models have been proposed to explain the photoquenching. Vincent, Bois, and Chantre[25] associate two states to EL2: one stable and optically active, the other metastable and optically inactive. When the center is in the metastable state, it is not sensitive to light. At a low temperature the center remains in its metastable state, provided that the thermal energy is not enough to overcome the energy barrier (0.35 eV) between the two states. Another model, by Levinson,[26] explains the photoquenching on the basis of an interaction between two or more defects forming a complex.

The measurement of the optical capture cross sections of deep levels usually provides good information about their physical nature. All the measurements of the electron optical capture cross section of EL2[15,16] previously reported have been performed at low temperatures, but the dependence of this magnitude on temperature has never been obtained. In our experimental work we have measured the thermal dependence of the electron optical capture cross section σ_n^0 of EL2. Thus we have obtained the photoionization spectra of EL2 at three temperatures covering a wide range limited at high temperatures by the thermal emission of EL2 and at low temperatures by the photoquenching. In the following section we describe the technique we have used for the experimental determination of the photoionization cross section of EL2 corresponding to the emission of electrons to the conduction band. This technique, called optical admittance spectroscopy,[27] is based on the measurement at low temperature of the conductance of a reverse-biased junction illuminated with a light of photon energy $h\nu$.

II. EXPERIMENT: OPTICAL ADMITTANCE SPECTROSCOPY

The optical admittance spectroscopy is a technique[27] which allows the measurement of the optical capture cross section of deep levels in junctions. It is based on the variations of the capacitance and conductance of a junction under illumination, as a function of the photon energy $h\nu$ and frequency ω of the measuring signal. We have derived an expression for the capacitance and conductance, as a function of frequency, of a reverse-biased junction containing a deep level. Thus, for a p^+n junction or a Schottky barrier on an n-type semiconductor having an acceptor level, we obtain the following expressions for the capacitance and the conductance:

$$C(\omega) = C_{\mathrm{HF}} + (C_{\mathrm{LF}} - C_{\mathrm{HF}})(e_n + e_p) \times \arctan[\omega/(e_n + e_p)], \tag{1}$$

$$G(\omega) = \frac{C_{\mathrm{LF}} - C_{\mathrm{HF}}}{2}(e_n + e_p)\ln[1 + (\omega/e_n + e_p)^2], \tag{2}$$

where $e_n = e_n^0 + e_n^t$ and $e_p = e_p^0 + e_p^t$. In these expressions C_{LF} and C_{HF} are the junction capacitance at low and high frequency, respectively, e_n^0 and e_p^0 are the optical emission rates for electrons and holes, respectively, and e_n^t and e_p^t are the thermal emission rates for electrons and holes, respectively.

Under illumination and at a temperature T low enough so that the thermal emission is negligible, the capacitance and conductance of a junction containing a deep level show an inflexion point and a maximum, respectively, when the photon energy $h\nu$ is scanned at a constant frequency of the measuring signal (Fig. 1). These capacitance and conductance variations are due to the change, relative to the frequency of the measuring signal, of the time constant of charge and discharge processes of the deep level around the y' point of the spatial charge region where the optical emission equals the thermal capture, i.e., $e_n^0(h\nu)$

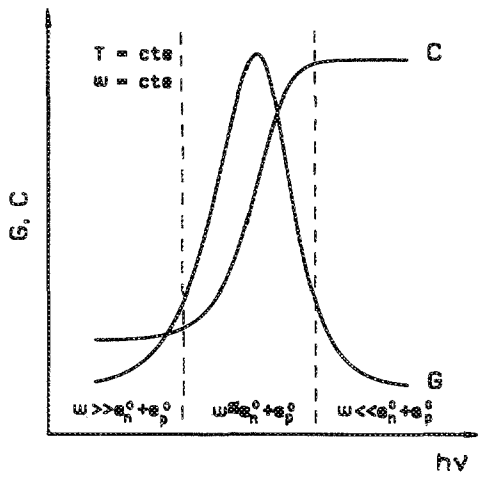

FIG. 1. Capacitance and conductance of a junction vs photon energy at a constant temperature and frequency.

$+ e_p^0(h\nu) = c_n n(y')$. c_n is the electron thermal capture rate, and $n(y')$ is the density of free electrons at the y' point. When the photon energy is such that $e_n^0(h\nu) + e_p^0(h\nu) \ll \omega$, the high-frequency capacitance and conductance of the junction are measured. When $e_n^0(h\nu) + e_p^0(h\nu) \gg \omega$, the low-frequency capacitance and conductance of the junction are measured. The inflexion point in the capacitance and the maximum in the conductance will occur at intermediate values of the emission rates, i.e., when $e_n^0 + e_p^0$ is approximately equal to the measuring signal frequency ω.

From Eq. (2) we deduce that the conductance as a function of the photon energy, $G(h\nu)$, at constant temperature and frequency, exhibits a maximum defined by the condition

$$e_n^0(h\nu) + e_p^0(h\nu) = \omega/1.98, \tag{3}$$

whose value $G(h\nu_m)$ verifies

$$G(h\nu_m) \propto \frac{e_n^0(h\nu_m)}{e_n^0(h\nu_m) + e_p^0(h\nu_m)} = \frac{1.98}{\omega} e_n^0(h\nu_m). \tag{4}$$

Therefore, the $G(h\nu)$ plots at constant T and ω enable us to determine the optical emission rates e_n^0 and e_p^0 as a function of $h\nu$ at the temperature T. Performing these measurements at different temperatures, we can obtain the thermal dependence of the optical emission rates. In the case where there are several deep levels in the gap, the total conductance will be the sum of the contributions of each optically active level. At a given frequency ω and photon energy $h\nu$, only those levels with optical emission rate $e_n^0(h\nu) + e_p^0(h\nu)$, close to the measuring signal frequency ω, will contribute to the conductance of the junction. The levels with an optical emission rate much higher or lower than ω will be in the low- or high-frequency condition, respectively, and have therefore no contribution to the total conductance of the junction. Consequently, a plot of the conductance as a function of the photon energy $h\nu$ will show a maximum for each optically active deep level located in the band gap. Thus we see that optical admittance spectroscopy is a true spectroscopic technique in the sense that it allows the optical emission rate of all deep levels located in the band gap of a semiconductor to be measured in a selective way.

In the case of the capacitance, the exact condition for the inflexion point of $C(h\nu)$, at a constant temperature and frequency, will depend upon the shapes of e_n^0 and e_p^0 as a function of $h\nu$, which is unknown *a priori*.

In the particular case of EL2, the electron optical emission rate e_n^0 is dominant at any photon energy, and this rate is the one we have obtained in our measurements.

Another important characteristic of this technique is that the measurements are performed in a small region around the y' point where the optical emission and thermal capture processes are balanced. The y' point is defined by the condition $e_n^0 + e_p^0 = c_n n(y')$, where c_n is the electron-capture thermal rate and $n(y')$ is the density of free electrons at the y' point. It is a point within the edge region of the spatial charge zone, close to the neutral region where the electric field is virtually zero. Consequently, the electric field at the y' point will be very small, and hence we can conclude that the optical admittance spectroscopy measures the optical emission rate at a point where the electric field is very small. The $\sigma^0(h\nu)$ spectra obtained will not depend upon the electric field, as is the case for other junction techniques in which the measured values are an average over the whole space-charge region and therefore depend upon the electric field existing in it. Thus the effects due to the strong electric fields, such as the Poole–Frenkel effect, can produce changes in the optical emission rates in a factor of 4 or higher relative to their values under no-electric-field conditions.

The optical emission rates can be written as the product of the optical capture cross section $\sigma_{n,p}^0$ times the photon flux $\Phi(h\nu)$:

$$e_{n,p}^0(h\nu) = \sigma_{n,p}^o \Phi(h\nu). \tag{5}$$

From this expression we see that, in order to measure the optical capture cross sections—which usually have very small values—it is necessary to use a very low-frequency ω and light flux Φ, which is as high as possible. In addition, very small optically generated currents require the use of a sensitivity electrometer. We used a model 617 Keithley meter.

Experimentally, the conductance measurement is carried out by measuring the in-phase component of the photocurrent relative to the measuring signal. Since the signals involved have very low frequencies, the synchronous detection can be performed numerically using a computer. When higher frequencies are required, as, for example, in thermal admittance spectroscopy, the use of analog synchronous detection systems, such as a lock-in analyzers, becomes necessary.

III. RESULTS AND DISCUSSION

We have used optical admittance spectroscopy to measure the optical capture cross section of the fundamental level of EL2. This level exhibits optical capture processes of photons in which electron emission to the conduction band dominates over hole emission to the valence band, at any photon energy $h\nu$. Therefore, the conductance measure-

ments yield the optical capture cross-section spectrum of electrons, $\sigma_n^0(h\nu)$.

The sample used was a Schottky barrier made on n-type ⟨110⟩ Czochralski GaAs which was boron implanted, resulting in a donor concentration $N_D = 9\times10^{16}$ cm^{-3}. An ohmic contact was made at the back by depositing and annealing at 500 °C a AuGe + Ni layer. The Schottky barrier was made with a 100-Å-thick semitransparent layer. Finally, a 2-μm-deep MESA etch was carried out. The resulting junction was circular, 800 μm in diameter. The EL2 concentration was determined from C-V measurements at high and low temperatures, yielding 8×10^{15} cm^{-3}.

We have measured the electron photoionization cross section spectrum $\sigma_n^0(h\nu)$ at three different temperatures covering the wide range in which optical admittance spectroscopy can be applied. This range is limited both at high and low temperatures. The high limit is the temperature at which thermal emission begins to be comparable to optical emission. DLTS data suggest that this happens at about 300 K. The lower limit is set by photoquenching which occurs at temperatures below 140 K and for photon energies between 0.9 and 1.35 eV, the range in which the EL2 center is insensitive to changes in illumination. This is why we have performed our experiments at 150, 200, and 270 K, thus covering the range mentioned above. Frequencies ranged from tens of milihertz up to several hertz. These low-frequency signals were obtained from an HP-3310-A signal generator. The light source was a Philips PF815R 1000-W tungsten filament halogen lamp covering the 0.3–2.5-μm wavelength range, equivalent to 4.1–0.4 eV of photon energy, enough to study the EL2 center. The monochromator used was a high-intensity Jobin-Yvon H25. Higher-order components were supressed by using suitable filters. The flux was measured with a PbS detector at room temperature.

Figure 2 shows several experimental curves of the conductance of the junction G/ω as a function of photon energy $h\nu$ for different frequencies and temperatures. In each curve we see three maximums. The first peak corresponds to a rising section in the $\sigma_n^0(h\nu)$ spectrum of the EL2 center and therefore to a change from high to low frequency. The second peak corresponds to a decreasing section in the spectrum (change from low to high frequency). The third peak is due to another rising section. Notice that these peaks exhibit different behaviors: The first two peaks are smoother and vary quite strongly with frequency and temperature, whereas the third one is sharper and its position hardly changes with frequency.

The position of the peaks allows us, using Eq. (3), to obtain the electron optical capture cross-section spectra $\sigma_n^0(h\nu)$ of the EL2 center at the three above-mentioned temperatures. The shape of the spectra is depicted in Fig. 3. In these spectra, the three different contributions corresponding to transitions from the EL2 level to the Γ, L, and X conduction-band minima of the GaAs can be seen. For photon energies $h\nu$ close to the band-gap energy the spectra rise again.

These experimental results were compared with the semiempirical model proposed by Chantre and co-workers.[15] In this model the optical capture cross section of a level with

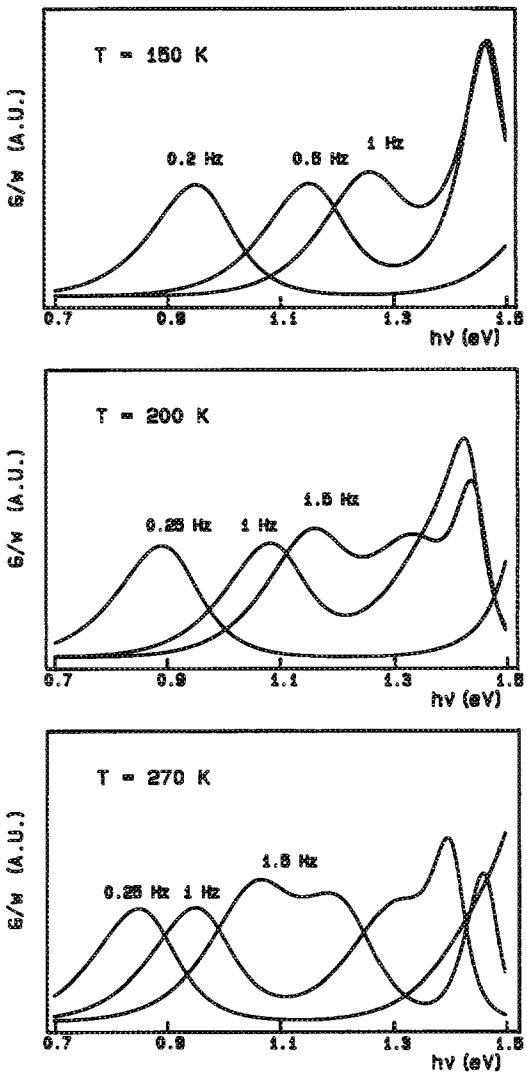

FIG. 2. Plots of G/ω vs photon energy for a GaAs Schottky barrier containing the EL2 center at $T = 150$, 200, and 270 K.

emission of electrons to the conduction band is the sum of three components, each of them due to the transition to each conduction-band minima from the EL2 level. A Dirac delta function is chosen for the potential at the impurity when the electron is trapped at the center. Four empirical parameters are thus introduced which can be fitted to the experimental results. The first parameter is the optical threshold energy E^0, which is the minimum photon energy necessary to excite optical electron emission and is related to the ionization energy of the center, E_T, through the Franck–Condon shift d_{FC} ($E^0 = E_T + d_{FC}$). The second adjustable parameter, α, accounts for the localization of the wave function of the electron trapped at the center. The other two adjustable parameters are the ratios P_L/P_Γ and P_X/P_Γ, where each P_i represents the amplitude of the lattice oscilations corresponding to a transition to each minimum. We used the experimental data from Fig. 3 for energies below 1.35 eV to obtain these parameters. We get an optical threshold energy of 0.86 eV, i.e., a Franck–Condon shift $d_{FC} = 0.10$ eV. The results for the other three empirical parameters, α, P_L/P_Γ, and P_X/P_Γ, for the three temperatures are given in Table I.

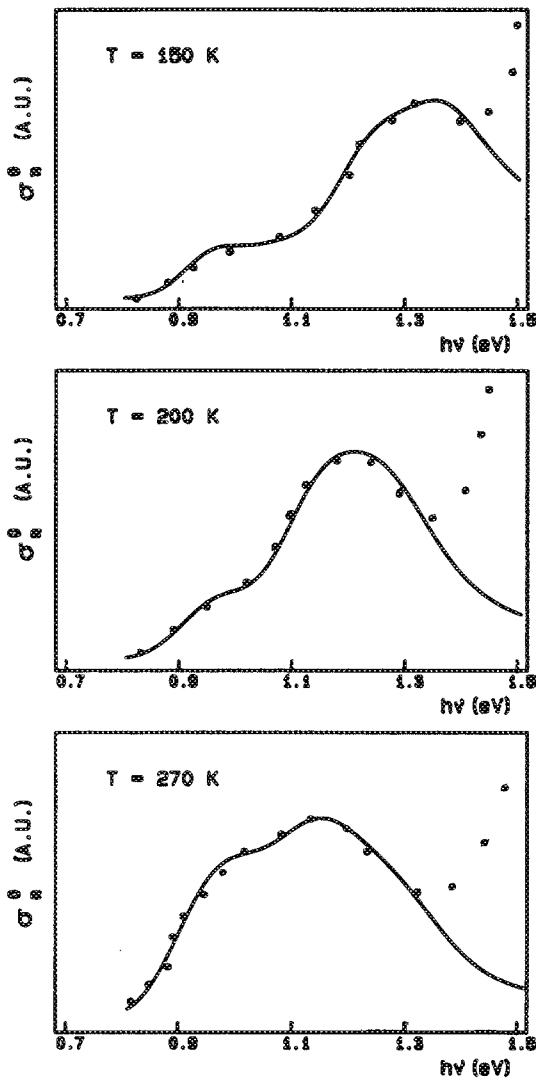

FIG. 3. Electron optical capture cross section σ_n^0 as a function of photon energy for the EL2 center in GaAs at $T = 150$, 200, and 270 K. The solid lines are the curves obtained using the semiempirical model of Chantre and co-workers (Ref. 15). The points (●) are our experimental results.

As can be seen from this table, these values are somehow scattered. This suggests that probably EL2 is not a single level as Chantre and co-workers' model assumes, but a family of very close levels as has been proposed by Taniguchi and Ikoma[8] and by Lagowski *et al.*[9] Taniguchi and Ikoma, in a more recent paper,[28] found that all these levels exhibit photoquenching, although the photoquenching efficiency varies from one level to another. From these results they conclude that all these levels must belong to the same family and hence have similar atomic structure. The transition from the stable to the metastable state is, however, different for each level, suggesting, therefore, the existence of different electron-lattice coupling modes. The existence of this family would confirm the cluster model proposed by Mochizuki and Ikoma.[7]

On the other hand, the rising section of the spectrum for photon energies $h\nu$ higher than 1.35 eV could be explained by assuming the existence of a shallow acceptor level located slightly above the valence band. This would agree with the model proposed by Hariu *et al.*[29] and derived from their

TABLE I. Values of the adjustable parameters obtained from experimental spectra using the semiempirical model of Chantre and co-workers (Ref. 15).

T (K)	α (Å^{-1})	p_L/p_Γ	p_X/p_Γ
150	0.085	0.25	0.25
200	0.04	0.5	0.1
270	0.03	0.1	0.025

quenching and recovery spectra of EL2 in semi-insulating GaAs measured by double-beam photoconductivity. These authors suggest that EL2 is formed from the association of the arsenic antisite As_{Ga} with an acceptor level. Their hypothesis is based on the general consideration that a given type of impurity level (donor in our case) tends to induce another one associated with the other type of defect or impurity (acceptor in our case) for energetic stabilization, particularly when the creation energy of the defect is less than the energy difference between the donor and acceptor levels. The existence of this shallow acceptor level might account for some experimental facts, such as the apparition of the P peaks in the OITS spectra, obtained by Morante *et al.*,[16] at photon energies close to the band-gap energy. This shallow level might also be related to the near-intrinsic quenching observed by Jiménez *et al.*,[23] which exhibits a maximum efficiency at an energy of 1.45 eV at 77 K.

IV. CONCLUSIONS

We have measured the optical electron capture cross section of EL2 in GaAs by optical admittance spectroscopy. This is a spectroscopic technique since the conductance of a junction containing several optically active deep levels shows a peak for each level when the photon energy is scanned. Another important feature of this technique is that it measures the optical capture cross section in a very small region around a point within the spacial charge zone very close to the neutral region and, therefore, at an electric field much lower than the maximum field. We have determined the electron photoionization spectra $\sigma_n^0(h\nu)$ at three different temperatures within a range limited at high temperature by thermal emission and at low temperature by photoquenching of EL2. We have fitted the experimental data to the semiempirical model of Chantre and co-workers,[15] which considers EL2 as a single defect emitting electrons to each GaAs conduction-band minima (Γ, L, and X). The resulting scattering in the values obtained for the empirical parameters suggest that, rather than a single level, EL2 seems to be formed by a family of very close levels located at midgap, as proposed by Taniguchi and Ikoma[8] and by Lagowski *et al.*[9] This family may be explained assuming that EL2 forms arsenic clusters, as suggested by Mochizuki and Ikoma.[7] The apparition of a rising section in the $\sigma_n^0(h\nu)$ spectrum for photon energies close to the band-gap energy can be explained by assuming that EL2 is formed by the association of arsenic antisite As_{Ga} with a shallow acceptor as has been suggested by Hariu *et al.*[29] This acceptor level would account for that section of the spectrum. This level may be due to the B_{As} or Ga_{As} defects, according to the data

reported by Dansas,[30] Bishop, Shanabrook, and Moore,[31] and Elliot,[32] or may be related with the carbon acceptor level according to the ESR results obtained by Elliot *et al.*[33] Other experimental results such as the P peaks in the OITS spectra obtained by Morante *et al.*,[16] as well as the near-intrinsic quenching observed by Jiménez *et al.*,[23] could also be explained by this hypothesis.

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