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Absolute photoionization cross sections of the acceptor state level of chromium in indium phosphide

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We have studied by deep-level transient spectroscopy and deep-level optical spectroscopy *n*-type chromium-doped InP. We definitively confirm that the $\text{Cr}^{2+}/\text{Cr}^{3+}$ acceptor state is positioned at $E_C - 0.4$ eV in InP. We have measured for the first time the absolute values of the photoionization cross sections of Cr^{2+} in InP by deep-level optical spectroscopy. The σ_n^0 cross section exhibits both a resonant and a nonresonant character. The former corresponds to the internal transition ${}^5T_2 \rightarrow {}^5E$ of Cr^{2+} , while the latter is attributed to the photoexcitation from the Cr^{2+} to the conduction band. The threshold of this transition at $E_C - 0.41$ eV indicates a very small Franck-Condon shift. The scales of the absolute photoionization values (σ_n^0) and photoneutralization (σ_p^0) cross sections towards the conduction and valence bands are the same, which seems to indicate no selection rules.

I. INTRODUCTION

The introduction of transition-metal ions as dopants into III-V compounds produces electronic properties which are of great interest from a theoretical point of view and useful from a practical one: the manufacturing of semi-insulating (s.i.) substrates of good quality used in planar technology in the fields of fast electronic and optoelectronic devices. The role of chromium in InP has been recently investigated by numerous techniques¹⁻⁶ in order to explain the compensation mechanism. Chromium doping produces semi-insulating material with resistivities of only about 10^4 Ω cm (by comparison, InP:Fe s.i. has a resistivity of 10^8 Ω cm).

Iseler¹ located by Hall and resistivity measurements a level associated with chromium at $E_C - 0.39$ eV in s.i. InP, assigning this level to the $\text{Cr}_{\text{In}}^{2+/3+}$ acceptor one. This result was confirmed by the photoconductivity measurements of Fung and Nicholas³: They located the $\text{Cr}^{2+/3+}$ acceptor level at $E_C - 0.4$ eV ($E_C - 0.47$ eV) at 300 K (4 K). On the other hand, the photoconductivity threshold found by Eaves *et al.*² leads to a very different conclusion.

In photoluminescence measurements, Koschel, Bishop, and McCombe⁴ observed a band peaking at 0.85 eV that they attributed to the internal transition ${}^5E \rightarrow {}^5T_2$ of the $\text{Cr}_{\text{In}}^{2+}$. The same interpretation has been given by Barrau *et al.*,⁵ who observed a zero-phonon structure at 0.886 eV associated with the 0.85-eV band. These results should locate the 5E excited state below the conduction-band minimum, and thus the chromium level lies around $E_C - 0.9$ eV, in agreement with Eaves *et al.* results² but in contradiction with the Iseler¹ and Fung and Nicholas³ results. Our first results⁶ using deep-level transient spectroscopy and deep-level optical spectroscopy techniques (DLTS-DLOS) were well interpreted by the chromium acceptor level at 0.4 eV below the conduction band, which was labeled IE3.

The aim of this paper is first to confirm definitively the position of the chromium acceptor level in InP and second to give absolute information on the spectrum of the photoionization cross sections σ_n^0 and σ_p^0 of IE3. The possibility of determining the absolute values of σ_n^0 and σ_p^0 is very important from a theoretical point of view. Indeed the photoionization cross sections can provide useful information, such as ionization energy of the level, coupling with the bands, and therefore some indications of the symmetry of defect wave functions.

In Sec. II, we give a description of the experimental technique used. The experimental results and discussion concerning InP:Cr are given in Secs. III and IV.

II. EXPERIMENTAL PROCEDURE

Samples come from a polycrystalline ingot, prepared by the conventional gradient freeze technique in a pyrolite boron nitride (PBN) crucible.⁷ The chromium concentration in the melt was 0.1% by weight, which is sufficient to reach the chromium solubility limit (2×10^{16} cm^{-3}) in the hot zone of the ingot. The studied samples come from the zone labeled *c* as defined in Ref. 7. Spark-source mass spectroscopy reveals silicon and chromium as the main impurities. The samples are *n* type ($\rho \sim 35$ Ω cm at room temperature). A doping profile measurement gives $N_d - N_a \simeq 6 \times 10^{14}$ cm^{-3} at 300 K. The experimental results reported below for $\text{Cr}^{2+/3+}$ in InP have been obtained using gold Schottky barriers of area 0.8 mm^2 on *n*-type InP. Before gold vaporization, first, the ohmic contacts are provided by small dots of Sn on the sides of the samples at 300 °C under hydrogen flow, and second, the surface is prepared by a standard cleaning and etching procedure followed by natural oxidation in dry atmosphere to increase the apparent Schottky barrier and thus to lower the leakage current of the diode under reverse bias conditions.⁸ The examined sample is mounted in a liq-

uid-N₂ cryostat or liquid-He cryostat, the window of this being made of CaF₂ and analyzed by DLTS and DLOS techniques.

DLTS measurements permit one to determine the apparent activation energy ΔE_a and the thermal capture cross section from the relation

$$\frac{e_{n,p}}{T^2} = 3.25 \times 10^{25} g_{n,p} \sigma_{\infty} \left(\frac{m_{n,p}^*}{m_0} \right) \exp\left(-\frac{\Delta E_a}{kT} \right), \quad (1)$$

where $e_{n,p}$ is the coefficient for thermal emission of carriers (electron or hole) from the trapping center, $g_{n,p}$ the degeneracy factor, and $m_{n,p}^*$ the electron (hole) effective mass at the minimum conduction (valence) band. The plot of $\log(e_{n,p}/T^2)$ as a function of $1/T$ is usually called the "signature" of the level.

For the DLOS experiment, the temperature of the sample is fixed so the thermal emission of carriers is neglected. The main feature of the DLOS technique is to use the fact that the derivative at time $t = 0$ of the photocapacitance transient, expressing the level occupancy in a reverse-biased junction capacitance, is directly related to $\sigma_n^0(h\nu)$ or $\sigma_p^0(h\nu)$ if initial conditions—all traps filled or empty—are fixed. These initial conditions are determined by various modes of operations—electrical, thermal or optical excitation—which depend on the trap characteristics and material type.⁹ For one electron trap of concentration N_T in an n -type material, the variation of charge due to the level at time $t = 0$ after a direct bias pulse to fill all the traps is

$$\left. \frac{dn}{dt} \right|_{t=0} = \sigma_n^0(h\nu) N_T \phi(h\nu). \quad (2)$$

$\phi(h\nu)$ is the photon flux at energy $h\nu$ per surface unity in the depletion layer. This charge variation leads to a variation of the capacitance value C of the Schottky given by

$$\frac{1}{C} \frac{dC}{dt} = \frac{1}{2n_e} \frac{dn}{dt} \left(1 - \frac{\lambda}{W} \right)^2, \quad (3)$$

with n_e the density of charge carriers, W the depletion zone width at reverse bias, and λ the zone width where the trap level is below the Fermi level.

From Eqs. (2) and (3) we have

$$\left. \frac{dC}{dt} \right|_{t=0} = \frac{CN_T}{2n_e} \left(1 - \frac{\lambda}{W} \right)^2 \sigma_n^0(h\nu) \phi(h\nu). \quad (4)$$

With the same capacitance measurement conditions, the DLTS peak of amplitude ΔC corresponding to the level gives

$$\frac{\Delta C}{C} = \frac{N_T}{2n_e} \left(1 - \frac{\lambda}{W} \right)^2. \quad (5)$$

From Eqs. (4) and (5) we have

$$\left. \frac{dC}{dt} \right|_{t=0} = \Delta C \sigma_n^0(h\nu) \phi(h\nu). \quad (6)$$

The DLOS signal is

$$S(h\nu) = \frac{1}{\phi(h\nu)} \left. \frac{dC}{dt} \right|_{t=0}. \quad (7)$$

From (6) and (7) we have

$$\sigma_n^0(h\nu) = \frac{S(h\nu)}{\Delta C}. \quad (8)$$

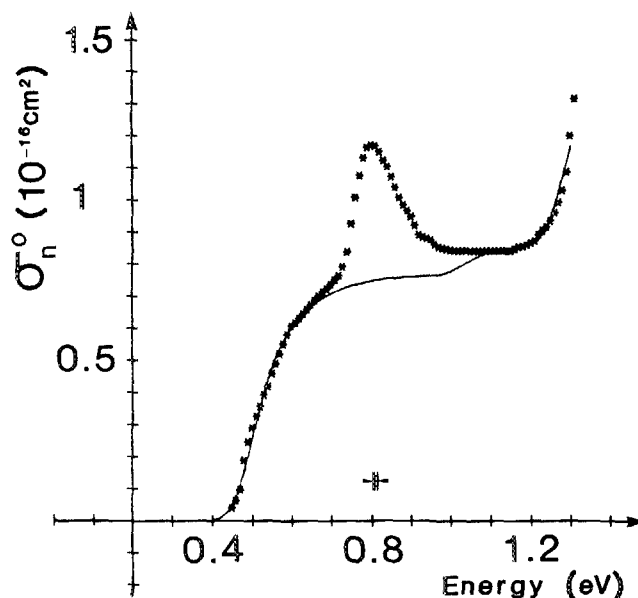


FIG. 1. Photoionization cross section $\sigma_n^0(h\nu)$ of the IE3 level obtained by electrical DLOS at $T = 89$ K. *, experimental points; —, fit of the nonresonant part of the photoionization cross section (see text).

To obtain the absolute value of $\sigma^0(h\nu)$, it is necessary to know the photon flux $\phi(h\nu)$ acting in the junction for the whole energy range [Eqs. (7) and (8)]. The DLOS apparatus has been already described.⁹ We use different monochromators for exciting light: a Perkin-Elmer 112B with a KBr of LiF prism, or a Bausch and Lomb high-intensity monochromator with 675-line/mm grating, blazed at $1 \mu\text{m}$ or

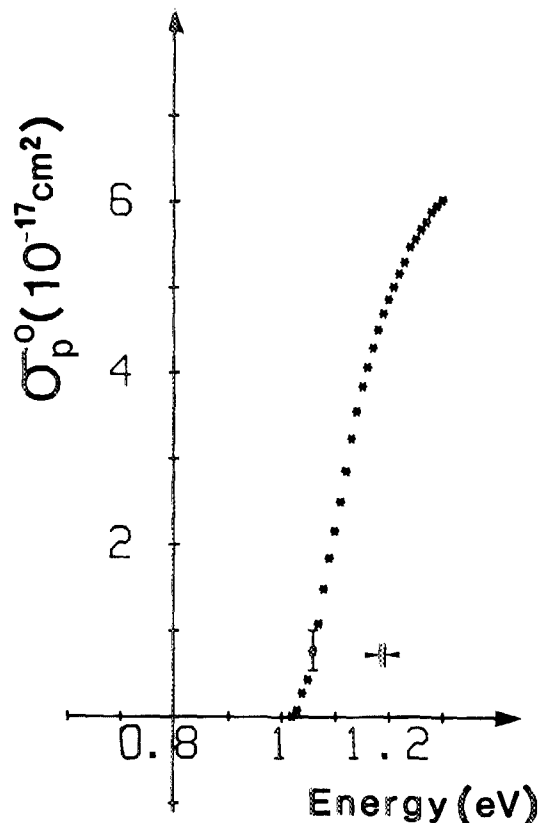


FIG. 2. Photoionization cross section $\sigma_p^0(h\nu)$ of the IE3 level obtained by thermal DLOS at $T = 157$ K.

337.5-line/mm grating blazed at $2\ \mu\text{m}$. The exciting sources that cover the entire energy range between 0.15 and 1.5 eV are Globar and tungsten light sources. The different cycles (electrical excitation, change of photon wavelength, temperature regulation, data acquisition and processing) are made by an on-line computer. The measure of the absolute value of the photon flux energy $\phi(h\nu)h\nu$ is performed by a sensitive pyroelectric detector Optilas KT3110, localized at the same position as the sample. Corrections are brought about following the experimental conditions. If the illumination is done on the side of the vaporized gold layer, the transmission was measured for this layer (semitransparent in this case) and taken into account for the exact value of $\phi(h\nu)$. If the illumination is done on the back side of the Schottky barrier, we use gold, mirrorlike, thick layers. The absolute measurements of σ_n^0 and σ_p^0 of the $\text{Cr}^{2+/3+}$ level in InP:Cr have been made over the entire gap energy range.

III. EXPERIMENTAL RESULTS ON InP:Cr

Two samples are intensively examined and give the same results. We observe in the samples only one main deep electron level ($\Delta E_n = 0.40\ \text{eV}$, $\sigma_\infty = 5 \times 10^{-15}\ \text{cm}^2$),

which has exactly the same thermal signature as the trap we have called IE3. In Ref. 6, we tentatively attributed this defect to the chromium acceptor level $\text{Cr}^{2+/3+}$ mainly because it was observed in *n*-type vapor-phase epitaxy (VPE) layers grown on s.i. substrates doped with Cr and it is well known that Cr can out-diffuse from the substrate to the epilayer. In the determination of the IE3 parameters, the following values are used: $g_n = 1$, $m_n^*/m_0 = 0.078$. The determined concentration of electrically active chromium is $3 \times 10^{14}\ \text{cm}^{-3}$.

The absolute optical cross sections $\sigma_n^0(h\nu)$ and $\sigma_p^0(h\nu)$ are given in Figs. 1 and 2. For σ_n^0 , we can observe an experimental onset at about 0.41 eV and a broad peak centered at 0.8 eV. This peak cannot be explained by the theory of the optical cross section for the transition between a deep level and the conduction band. We give in Fig. 1, for illustration, the best fit given by the phenomenological model we used to analyze the optical cross sections of the deep levels in III-V compounds.⁹ This analysis has been made in details for the deep Cr level in GaAs.¹⁰

The fit of Fig. 1 corresponds to the following values of the model parameters:

Binding energy E_n	E_n^0	d_{FC}	P_Γ	P_L	P_X	α^{-1}
0.4 eV	0.475 eV	75 meV	1	0.0065	0.064	0.5 Å

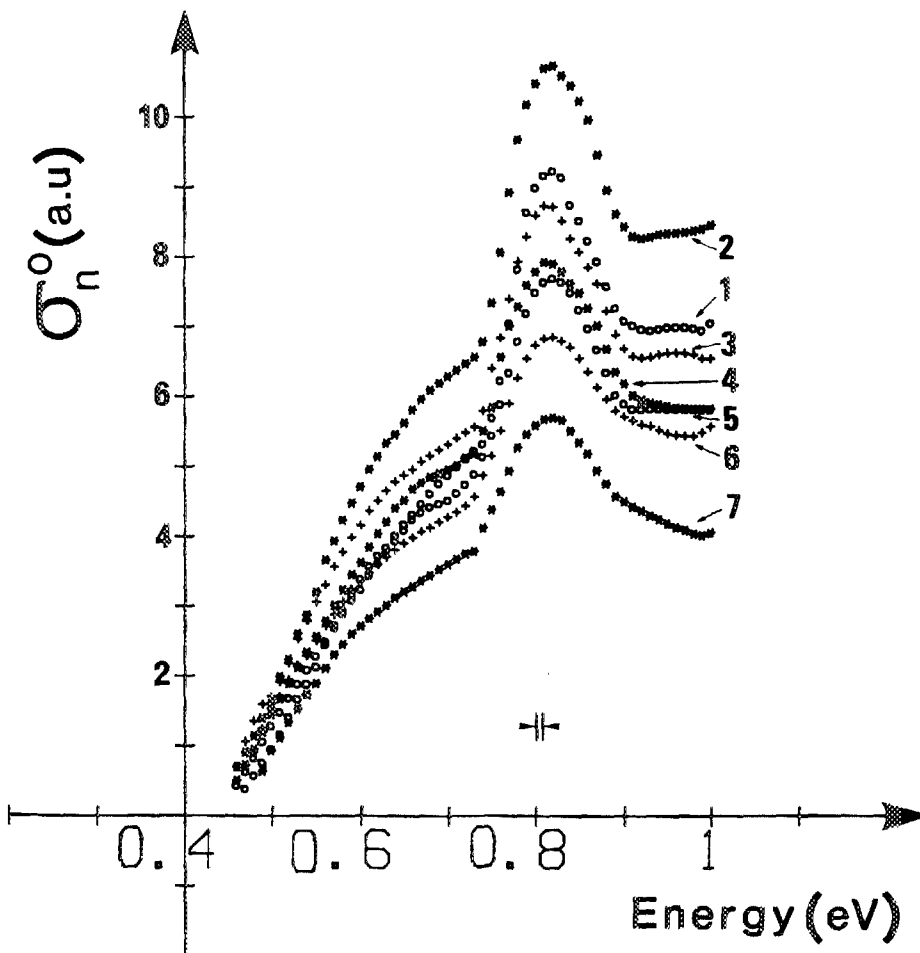


FIG. 3. Photoionization cross sections $\sigma_n^0(h\nu)$ of the IE3 level at different temperatures. 1, $T = 9\ \text{K}$; 2, $T = 33\ \text{K}$; 3, $T = 63\ \text{K}$; 4, $T = 80\ \text{K}$; 5, $T = 99\ \text{K}$; 6, $T = 120\ \text{K}$; 7, $T = 136\ \text{K}$.

Here E_n^0 is the optical ionization energy, d_{FC} is the Franck-Condon energy ($E_n^0 = E_n + d_{FC}$), P_Γ , P_L , and P_X are weighting factors adjusted in order to fit the transitions towards the Γ , L , and X bands, respectively, and α^{-1} gives information on the wave-function extension of the bound electron. In the mathematical formulation,⁹ we consider the electronic transitions allowed and the deep trap potential as a Dirac one.

Figure 3 shows σ_n^0 at different temperatures. The half-width of the 0.8-eV band can be classically analyzed, considering this one as Gaussian.¹¹ The shape of the band is then given by

$$\sigma_G^0 = B \exp\left(-\frac{(h\nu - E_0)^2}{2\sigma^2}\right), \quad (9)$$

where B is the normalization parameter, E_0 is the energy of the maximum, and

$$\sigma = \sigma_0 \left(\coth \frac{\hbar\omega}{2kT}\right)^{1/2}, \quad (10)$$

$$\sigma_0 = S^{1/2} \hbar\omega. \quad (11)$$

We use generally the value of the half-width:

$$W(T) = 2\sigma(2 \ln 2)^{1/2}. \quad (12)$$

In these expressions, the symbols have their usual meaning, S being the Huang-Rhys factor and $\hbar\omega$ the effective phonon energy which is coupled to the level. In the fundamental state, the Franck-Condon shift is given by

$$d_{FC} = S\hbar\omega.$$

The results of the best fit (Fig. 4) are

$$W(0 \text{ K}) = 79 \pm 2 \text{ meV},$$

$$\hbar\omega = 15 \text{ meV},$$

$$S = 5,$$

so

$$d_{FC} = S\hbar\omega = 75 \text{ meV}.$$

IV. DISCUSSION

A. Evidence of the $\text{Cr}^{2+/3+}$ acceptor level

Following our DLTS measurements on bulk samples InP:Cr and the VPE layer on a Cr-doped substrate, we can claim without ambiguity that IE3 is clearly connected with the presence of chromium. The photoionization cross section σ_n^0 of IE3 shows a very characteristic resonance at 0.8 eV, the bottom of that band being at 0.76 eV. This latter transition has been recently observed by Clerjaud *et al.*¹² in optical absorption. The zero-phonon line of that band is located at 0.756 eV with a fine structure typical of the internal transition of Cr^{2+} in III-V compounds.¹³ This fact means that in our experiment we perform photoionization of Cr^{2+} in its ground state 5T_2 . As a consequence the threshold at 0.41 eV gives the energy level of $\text{Cr}^{2+/3+}$ in agreement with other authors.^{1,3,14,15} The excited state 5E is the conduction band. So, as it is shown by the analysis of σ_n^0 , we can distinguish for σ_n^0 two kinds of transitions, namely the internal and

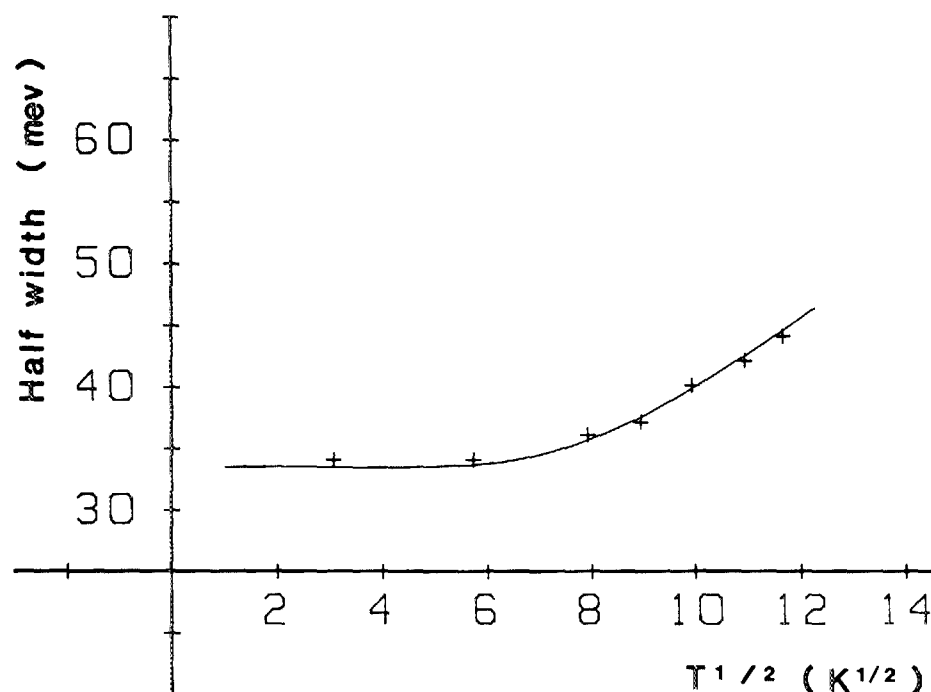


FIG. 4. Temperature dependence of the 0.8-eV band half-width $W(T)$. +, experimental points; —, best fit [Eq. (12) with $W(4 \text{ K}) = 79 \text{ meV}$, $\hbar\omega = 15 \text{ meV}$, $S = 5$].

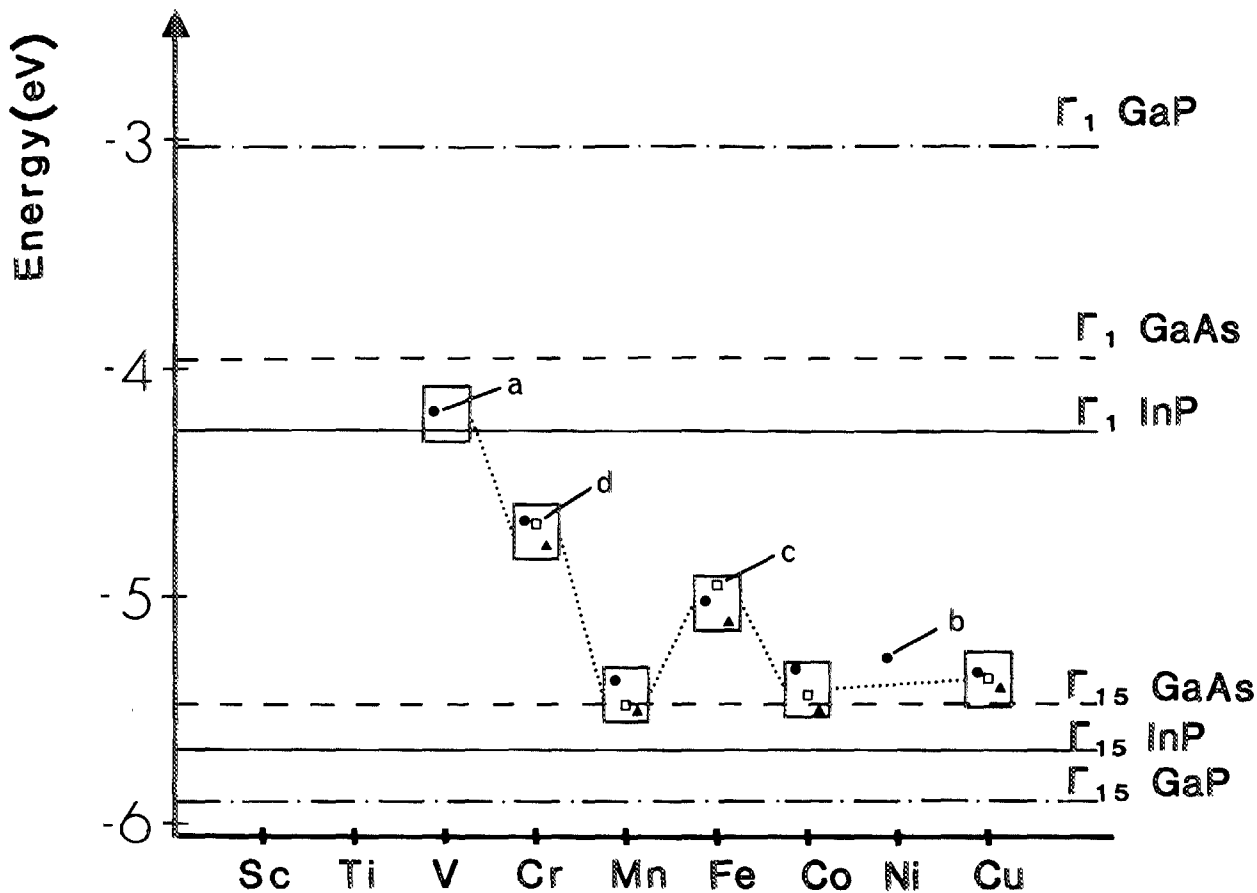


FIG. 5. Acceptor states (M^{2+}/M^{3+}) for the 3d transition metal in GaAs, InP, and GaP related to vacuum. ●, GaAs; □, InP; ▲, GaP. *a* indicates data from Ref. 20; *b* from Ref. 21; *c* from Ref. 22; *d*, this work. Other data are from Ref. 19.

the photoionization transitions. The former is written in our context

$${}^5T_2 + h\nu \rightarrow {}^{2S+1}\Gamma, \quad (13)$$

where S and Γ are labels of the final state. These excitations are currently described in the framework of the crystal field¹⁶ or molecular scheme.¹⁷ The photoionization transition is written

$${}^5T_2 + h\nu \rightarrow {}^{2S+1}\Gamma + e^- \text{ (in the conduction band)}. \quad (14)$$

The final state is that of Cr^{3+} with an electron going away. As usual we have supposed that Cr^{3+} is its ground state 4T_1 , but Cr^{3+} can be in an excited state in the final state of the transition.¹⁸ The use of a Dirac potential in the mathematical formulation⁹ for the best theoretical fit of the photoionization transition is in agreement with an acceptor state.

B. Trends in level positions

It is interesting to compare the thermal ionization energy of the Cr^{2+} charge state (0.4 eV) in InP with the other III-V compounds: GaAs and GaP. It is expected that a deep level related to a given transition metal is mainly tied to the vacuum and independent of the host lattice in III-V compounds.¹⁹ We have reported in Fig. 5 the well-established ionization energies of the acceptor state related to chromium

on an absolute energy scale and the principal band edges. The absolute impurity level for $\text{Cr}_{\text{In}}^{2+/3+}$ is well linked to the vacuum. Moreover, the energy position of the $\text{Cr}_{\text{In}}^{2+/3+}$ acceptor state level in InP is consistent with the general trend of the deep-level positions of transition elements in III-V compounds.²³

C. Transition ${}^5T_2 \rightarrow {}^5E$

The band at 0.8 eV corresponds to the internal transition ${}^5T_2 \rightarrow {}^5E$ of Cr^{2+} ; it is interesting to make a comparison with the same transition in GaAs (see Table I). If we neglect the electron phonon coupling in the excited state 5E , we can interpret the Franck-Condon shift as a result of the Jahn-Teller coupling in the ground state 5T_2 . So, the two systems exhibit the same Jahn-Teller energy (~ 70 meV).

Moreover, the effective mode energies clearly depend on the methods that have been used by different authors. Some of them use the separation $S\hbar\omega$ between the zero-phonon line and the maximum of the band together with the ratio e^{-S} between the area of the zero-phonon line and the area of the full band. Other authors, as we did, extract the effective mode energy from the dependence of the width on temperature.

We believe that the discrepancy between the two methods does not come from some error inherent to experiment

TABLE I. Comparison of some characteristics of the internal transition ${}^5T_2 \rightarrow {}^5E$ for Cr^{2+} in GaAs and Cr^{2+} in InP.

	Transition	$\hbar\omega_{eg}$ (meV)	S	$S\hbar\omega_{eg}$ (meV)	References
Cr^{2+} in GaAs	0.820	15	5	75	10
Cr^{2+} in GaAs	0.820	9	8.5	75	18
Cr^{2+} in InP	0.756	15	5	75	Present work
Cr^{2+} in InP	0.756	~ 8.7	7	~ 60	12

but rather is due to the oversimplified models. As usual we have supposed that the coupling to one effective mode works. In fact, the center is coupled to many modes. So a possible explanation seems to be the breakdown of the approximation of single-mode coupling.

D. Comparison between σ_n^0 and σ_p^0 optical cross sections

In the widely employed crystal-field picture,¹⁶ the optical transitions from a $3d$ trap state to the s -like conduction band are assumed to be parity-forbidden, whereas transitions from the p -like valence band to the trap are allowed. If we look to the nonresonant part of the photoionization cross section σ_n^0 (Fig. 1) we note that (i) the absolute values of the transition towards the conduction band (σ_n^0) and towards the valence band (σ_p^0) are both of the order of 10^{-16} cm^2 , and (ii) for the best fit of the nonresonant part of σ_n^0 , we consider the electronic transitions allowed in the mathematical formulation.

Very recent experimental data on Cr in ZnS (Ref. 24) show the optical cross sections σ_n^0 and σ_p^0 to be both of the order of 10^{-17} cm^2 . So, as a consequence no selection rules seem to work. Recent theoretical calculations of Vogl and Baranowski²⁵ show that the ratio σ_n^0/σ_p^0 is 1.23 for Cr in CdSe, in quantitative agreement with the data for ZnS:Cr and our results for InP:Cr. This result reflects the extended hostlike character of the trap wave function which strongly couples to both s - and p -type Bloch states and does not favor the model of a weak crystal field scheme.

In the case of Cr^{2+} , t_2 orbitals of the fundamental state (5T_2) are partly hybridized with ligand orbitals,²⁵ and transitions to the two bands have the same order of magnitude in contrast to what is observed for the vanadium donor level in InP ($V^{3+/4+}$),²⁶ where the fundamental state has e orbitals.

V. CONCLUSION

With our DLOS technique, we have experimentally shown the correlation between the onset at 0.41 eV and the internal transition ${}^5T_2 \rightarrow {}^5E$ of Cr^{2+} . This last fact allows us to locate the $\text{Cr}^{2+}/\text{Cr}^{3+}$ level 0.4 eV below the conduction band.

We have measured the absolute cross sections of photoionization of Cr^{2+} . We find similar values for both transitions to the conduction band and to the valence band. This result cannot be explained by a simple model of a parity-allowed $\text{Cr}^{3+} \rightarrow \text{Cr}^{2+}$ process (from p -type valence band to the d shell of Cr) and a parity-forbidden $\text{Cr}^{2+} \rightarrow \text{Cr}^{3+}$ process (from the d shell of Cr to the s -like conduction band). This effect can be ascribed to a strong hybridization of the impurity $3d$ function with host wave functions.

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