Optically induced nuclear magnetic field in InP

W. Farah, M. Dyakonov,* D. Scalbert, and W. Knap
Groupe d'Etude des Semiconducteurs, UMR 5650 CNRS, Université Montpellier 2, Place Eugène Bataillon, 34095 Montpellier Cedex, France

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We report on an optical manifestation of dynamic nuclear-spin polarization in optical pumping of *p*-InP. The electron-spin orientation is measured via the circular polarization degree of the luminescence. Depolarization by a transverse magnetic field (Hanle effect) yields the values of the electron lifetime and spin relaxation time. Dynamic nuclear polarization manifests itself when longitudinal and transverse magnetic fields are applied simultaneously. Optically induced nuclear magnetic fields up to 680 G are observed. All the experimental polarization curves can be well explained if one assumes that the nuclear polarization depends on the total external magnetic field. A qualitative explanation of this unexpected result is proposed.
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I. INTRODUCTION

The electrons created in the conduction band of a semiconductor in the process of interband absorption of circularly polarized light are spin oriented, and this spin orientation is easily detected by measuring the degree of circular polarization of the luminescence. Moreover, due to hyperfine interaction with the lattice nuclei, the electron-spin polarization is transferred to the nuclear-spin system, resulting in an appreciable nuclear polarization. In turn, the nuclear polarization acts back on the electron spins and thus modifies the polarization of the luminescence. Starting with the pioneering work of Lampel,[1] a large body of experimental and theoretical work has been done on optical spin orientation of electrons and nuclei in semiconductors (for a review, see Refs. 2–5).

Optical manifestations of dynamic nuclear polarization, including optical detection of nuclear magnetic resonance, in semiconductors were first demonstrated experimentally by Ekimov and Safarov[6] (see also Ref. 7) in studies of the polarization of the luminescence in a longitudinal (with respect to the pumping light beam) magnetic field. Later, Dyakonov *et al.*[8] showed that the so-called oblique Hanle effect is an universal tool for optical detection of nuclear polarization under optical spin pumping in semiconductors. This technique was utilized in a number of works, and spectacular nuclear effects were observed, especially at very low fields, on the order of 1 G.[3,4]

The back action of the polarized nuclei on the electron spin system is very well explained by the concept of the effective nuclear magnetic field, introduced in Ref. 9. Nuclear fields of about 2 kG were measured in $Ga_{1-x}Al_xAs$,[6,7] and fields on the order of tens of kG could be expected under favorable conditions.[10] It was also shown[11] that the appearance of nuclear polarization is in fact the result of a deep cooling of the nuclear-spin system by the nonequilibrium spin-oriented electrons. It should be noted that, although the general features of electron-nuclear-spin interactions under optical spin orientation in semiconductors are fairly well established, the details are not at all clear. It is known that localization of electrons by donors greatly enhances the rate of nuclear polarization, and that nuclear-spin diffusion may play a major role, as was clearly demonstrated experimentally by Paget.[12] However, the dependence of the nuclear polarization rates and leakage factors on sample doping, temperature, magnetic field, and excitation power was never investigated systematically, and remains unknown.

The nuclear- and electron-spin systems are strongly coupled, and typical nonlinear effects, like hysteresis and slow self-sustained oscillations of the luminescence polarization, were observed in optical spin-pumping experiments.[3] One such nonlinear effect is the dynamic nuclear self-polarization phenomenon predicted in Ref. 13 (see also Refs. 2 and 14), but not yet observed experimentally. The prediction is that if the nonequilibrium electrons are maintained in a disordered spin state, which may be done by interband absorption of nonpolarized or linearly polarized radiation, a substantial nuclear polarization must spontaneously arise. Its appearance has the features of a second kind of phase transition with a critical crystal temperature T_c. The effect should manifest itself by an appearance of circularly polarized luminescence at $T<T_c$, when the excitation is not polarized. (A similar effect in an optically pumped atomic vapor was later considered theoretically,[15] and recently observed experimentally.[16]) The critical temperature depends on the strength of the hyperfine interaction between the electrons and the nuclei, as well as on the nuclear leakage factor. The critical temperature also depends on the ratio τ_s/τ, where τ_s is the electron-spin relaxation time and τ is the lifetime. For favorable conditions (small leakage and large τ_s/τ ratio) the critical temperature for heavy nuclei should be on the order of several K.

The purpose of the present work is to study dynamic nuclear polarization under optical spin pumping in InP with the use of the oblique Hanle effect. This material was chosen because the high value of the hyperfine constant for the In nucleus allows one to expect the appearance of large optically induced nuclear fields and pronounced nuclear effects in the polarization of the luminescence. An optimistic estimate of the critical temperature for nuclear self-polarization in InP gives $T\approx 5$ K. The first optical spin pumping experiments in InP were done in Ref. 17. Optical detection of electron-spin resonance and determination of the electron *g*

factor by the spin orientation method were reported in Ref. 18. However so far no nuclear effects were seen in this material.

II. THEORETICAL BACKGROUND

A. Nuclear magnetic field produced by optical spin pumping in semiconductors

Hyperfine interaction plays a dual role. On the one hand, it leads to nuclear-spin relaxation by electrons and dynamic nuclear polarization, which occurs if the electron-spin system deviates from equilibrium, as is the case in optical spin pumping experiments. On the other hand, in the presence of nuclear polarization, it leads to the appearance of an effective magnetic field $\mathbf{B}_N$, acting on the electronic spins.

The nuclear-spin-lattice relaxation time T_1 at low temperatures is normally several orders of magnitude longer than the time T_2, on the order of 10^{-4} s, characteristic of spin-spin interactions within the nuclear-spin system. Thus this system is to a large extent isolated from the lattice. Owing to internal interactions, during a time of the order of T_2, it reaches thermodynamic equilibrium with a spin temperature, which may differ from the lattice temperature.[19] The rate of equilibration of the spin and lattice temperatures is defined by the long time T_1.

In the presence of an external field B, the average nuclear spin $\mathbf{I}_{\mathrm{av}}$ is given by the usual thermodynamic formula[19]

$$\mathbf{I}_{\mathrm{av}}=(I+1)\mu_I B/(3\theta), \tag{1}$$

where I and μ_I are the values of the nuclear spin and magnetic moment, and θ is the nuclear-spin temperature in units of energy. This formula is valid in the high-temperature approximation $\mu_I B \ll \theta$.

Thus the dynamic nuclear polarization by photoexcited spin-oriented electrons is due to the cooling of the nuclear-spin system. The steady-state spin temperature of the nuclear system interacting with spin-polarized nonequilibrium electrons is given by the expression[11]

$$\frac{1}{\theta}=\frac{4I}{\mu_I}\frac{\mathbf{B}\cdot\mathbf{S}}{B^2+\widetilde{B}_L^2}, \tag{2}$$

where $\mathbf{S}$ is the mean value of the electron-spin vector, the quantity $\widetilde{\mathbf{B}}_L$ is of the order of the local magnetic field produced at a nuclear site by neighboring nuclei.[19]

The concept of the effective nuclear magnetic field $\mathbf{B}_N$, introduced in Ref. 9, is very well justified for electrons in semiconductors, since the volume occupied by a free (or bound) electron is defined by its de Broglie wavelength (or the localization radius), and this volume is much greater than the unit-cell volume, i.e., it includes many lattice nuclei. The nuclear field $\mathbf{B}_N$ may be written as

$$\mathbf{B}_N=A\mathbf{I}_{\mathrm{av}}/(\mu g)=b_N\mathbf{I}_{\mathrm{av}}/I, \tag{3}$$

where A is the hyperfine constant; μ is the Bohr magneton; g is the electron g factor; and

$$b_N=\frac{16\pi}{3gv_0}\mu_I\eta\xi \tag{4}$$

is the maximal possible value of the nuclear field, obtained for 100% nuclear polarization, when $\mathbf{I}_{\mathrm{av}}=I$, ν_0 is the unit cell volume, ξ is the number of nuclei of the species considered in the unit cell, $\eta=|u(0)|^2$, and $u(0)$ is the electron Bloch amplitude at the site of the nucleus. [The Bloch function $u(\mathbf{r})$ is assumed to be normalized to the volume of the unit cell, so that η is a dimensionless parameter.]

Equations (1)–(3), combined, result in the following expression for the nuclear field:[11]

$$\mathbf{B}_N=\tfrac{4}{3}b_N(I+1)\frac{(\mathbf{B}\cdot\mathbf{S})\mathbf{B}}{B^2+\widetilde{B}_L^2}. \tag{5}$$

Thus the nuclear magnetic field is always directed along the external field $\mathbf{B}$, or opposite to it, depending on the sign of the spin temperature (the sign of $\mathbf{B}\cdot\mathbf{S}$). If $\mathbf{S}$ is perpendicular to $\mathbf{B}$, the nuclear field is absent. Again, Eq. (5) is valid in the high-temperature approximation, when $B_N \ll b_N$. In the derivation of this equation, it was also assumed that the hyperfine interaction with nonequilibrium spin-polarized electrons is the dominant interaction which defines the nuclear-spin temperature, or, in other words, that the nuclear relaxation time T_{1e}, due to interaction with spin-polarized electrons, is much shorter than the time T_1 describing other spin relaxation processes. If these two times are comparable, an additional ''leakage factor'' $f=T_1/(T_1+T_{1e})$, should be introduced into the right-hand side of Eqs. (2) and (5).

In the following we will neglect the term $\widetilde{B}_L^2$ in Eq. (5), since we will not be concerned with the region of very small magnetic fields $B \approx \widetilde{B}_L$ (on the order of several G), where it may be important.

With the nuclear field present, the total magnetic field acting on the electron spins will be equal to $\mathbf{B}+\mathbf{B}_N$, and this is why the nuclear polarization may manifest itself in optical polarization phenomena. In order to evaluate the role of the nuclear field we should estimate the value of b_N in Eq. (5), using Eq. (4).

There are two isotopes of In, In^{113} and In^{115}, with natural abundances 4.3% and 95.7%, respectively, spin $\frac{9}{2}$, and values of the magnetic moment μ_I equal to 5.523 and 5.534, in units of the nuclear magnetic moment. The P^{31} nucleus has spin $\frac{1}{2}$ and magnetic moment 1.132.[20] The η values are not known for InP. For In we will use the value $\eta=6300$ measured by Gueron[21] in InSb. Presumably, for the P^{31} nucleus the value of η is much smaller, since it is generally known that the η values increase greatly for heavy nuclei. Thus, with good accuracy, we can discard altogether the small contribution of P^{31} to the nuclear field b_N. We now calculate b_N with the use of Eq. (4), assuming that η/ν_0 (i.e., the electron wave function at the nuclear site) for the In nucleus has the same value in InP as in InSb.[22] We then obtain

$$b_N=34.4 \text{ kG}. \tag{6}$$

In the absence of appreciable leakage due to spin-lattice relaxation, the critical temperature T_c for the nuclear self-polarization phase transition under optical pumping by unpolarized light is related to b_N by the equation[13] $kT_c=(I+1)\mu g b_N/3$, which gives $T_c=5.3$ K.

B. Influence of nuclear polarization on the Hanle effect in an oblique field

In semiconductors with the band structure as in GaAs, or InP, the degree of circular polarization of the interband luminescence, P, is equal to the projection of the average electron spin on the direction of the observation, S_z. If the magnetic field makes an angle ϑ with this direction, the polarization of luminescence, $P(B)$, under stationary conditions, is given by the formula[2]

$$\frac{P(B)}{P(0)}=\cos^2\vartheta+\frac{\sin^2\vartheta}{1+(B/B_H)^2}, \tag{7}$$

where

$$P(0)=\frac{1}{4}\,\frac{\tau_s}{\tau+\tau_s} \tag{8}$$

is the stationary value of polarization in zero field, τ and τ_s are the electron lifetime and the spin relaxation time respectively. In Eq. (7), B_H is a characteristic field determined by the relation

$$B_H=\frac{\hbar}{\mu g}\left(\frac{1}{\tau}+\frac{1}{\tau_s}\right). \tag{9}$$

Equation (7) describes the so-called oblique Hanle effect, B_H being the half-width of the depolarization curve. For $\vartheta=90°$, it gives the normal Hanle effect.

If the nuclear field is built up, it should be added to the external magnetic field. Thus in Eq. (7) we should replace $\mathbf{B}$ by $\mathbf{B}+\mathbf{B}_N$, with $\mathbf{B}_N$ given by Eq. (5). In order to evaluate the scalar product $\mathbf{B}\cdot\mathbf{S}$, entering Eq. (5), we write down the equation describing the mean electron-spin vector $\mathbf{S}$ under stationary conditions[2]

$$\frac{\mu g}{\hbar}\,\mathbf{S}\cdot(\mathbf{B}+\mathbf{B}_N)+\mathbf{S}\left(\frac{1}{\tau}+\frac{1}{\tau_s}\right)=\frac{\mathbf{S}_0}{\tau}, \tag{10}$$

which takes into account the spin precession in the total magnetic field $\mathbf{B}+\mathbf{B}_N$, spin relaxation and recombination, and spin pumping. $\mathbf{S}_0$ is a vector directed along the pumping light beam (z axis), with an absolute value $\frac{1}{4}$. We now take the scalar product of the left- and right-hand sides of this equation with $\mathbf{B}$, and, since $\mathbf{B}_N$ is directed along $\mathbf{B}$, we obtain

$$\mathbf{B}\cdot\mathbf{S}=\frac{\tau_s}{\tau_s+\tau}\,\mathbf{B}\cdot\mathbf{S}_0=B_zP(0). \tag{11}$$

If $\mathbf{B}$ is perpendicular to the pumping light beam (normal Hanle effect), then $\mathbf{B}\cdot\mathbf{S}=0$, and the nuclear polarization is not built up. This is the reason why, for studies of nuclear effects, the oblique Hanle effect should be used.

Replacing $\mathbf{B}$ by $\mathbf{B}+\mathbf{B}_N$ in Eq. (7), and using Eqs. (5) and (11), we may now obtain the final expression for the magnetic-field dependence of the degree of circular polarization of the luminescence. In doing this, for convenience of interpretation of our data, we introduce as variables the two projections of the external magnetic field, $B_\parallel=B_z$ (along the pumping beam of light), and $B_\perp$ (perpendicular to the beam), rather than the angle ϑ and the absolute value of the field B.

We also introduce the leakage factor f in Eq. (5) and neglect $\widetilde{B}_L^2$, as explained above. The result is

$$\frac{P(\mathbf{B})}{P(0)}=1-\frac{x^2}{x^2+z^2+\left(1+\dfrac{\alpha z}{x^2+z^2}\right)^{-2}}, \tag{12}$$

where $z=B_\parallel/B_H$ and $x=B_\perp/B_H$ are the parallel and perpendicular magnetic-field components in units of the Hanle linewidth B_H, and the parameter α, related to the nuclear magnetic field, is given by the formula

$$\alpha=\frac{4}{3}\,(I+1)P(0)f\,\frac{b_N}{B_H}. \tag{13}$$

The most interesting feature of Eq. (12) is its asymmetry in z, i.e., for a given sign of the circular polarization of the pumping light, the polarization of luminescence depends on whether the angle ϑ between the external field and the pumping beam is less or greater than 90°. This asymmetry obviously comes from the factor $\mathbf{B}\cdot\mathbf{S}$ in Eq. (5). Depending on the sign of this scalar product, the nuclear field $\mathbf{B}_N$ is directed along the external field $\mathbf{B}$ or opposite to it. In the latter case the nuclear field may compensate the external one, so that, at some $\mathbf{B}$, $P(\mathbf{B})=P(0)$. As can be seen from Eq. (12), this happens if $\alpha z<0$ at

$$x=(|\alpha z|-z^2)^{1/2}, \tag{14}$$

provided that $|z|<|\alpha|$. [The sign of α depends on the sign of $P(0)$. In the following we will consider α as positive.] Thus, for a given z, which satisfies this inequality, the positions of the maxima of $P(\mathbf{B})$, as a function of x, give directly the value of the parameter α.

III. EXPERIMENT

For moderate optical excitation levels, efficient electron-spin polarization can be obtained only in p-type samples.[2] We used commercially available p-type InP bulk samples. They were doped with Zn at a level of 4×10^{15} cm^{-3}. In all optical experiments,[9] mechanically and chemically polished (100) surface was used. The experimental setup that we used was a conventional apparatus for luminescence measurements, with polarization elements to produce or to analyze the circular polarization.[5] The luminescence was excited by a tunable Al_2O_3:Ti^{2+} laser pumped by an argon laser. The excitation energy was varied between 1.426 and 1.438 eV, slightly above the band gap.

The excitation laser intensity was modulated at 400 Hz by a mechanical chopper. A photoelastic $\lambda/4$ modulator working at a frequency of 50 kHz was used in two different configurations:

In the first configuration (a), the modulator was placed on the laser beam. It served to modulate the laser polarization between σ^+ and σ^-, while polarization of the luminescence was analyzed with a fixed circular polarizer. Such a configuration avoids nuclear polarization, which responds much slowly compared to the modulation frequency.

In the second configuration (b), the modulator was placed before the entrance slit of the spectrometer and followed by a linear polarizer. It served to analyze the circular polarization of luminescence excited in this case with a fixed σ^+ or σ^- polarization.

Photoluminescence was detected by a GaAs photomulti-

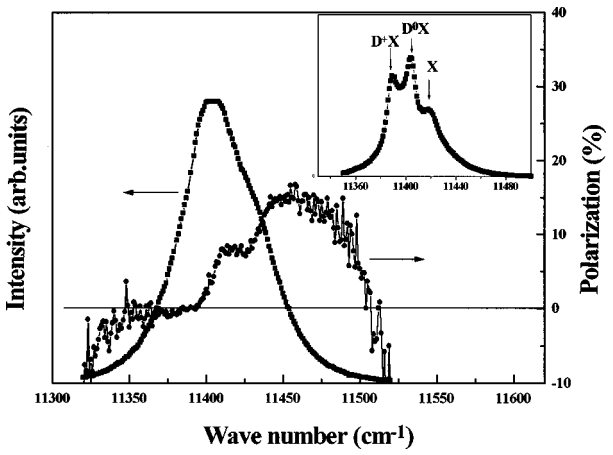

FIG. 1. Intensity (solid squares) and circular polarization degree (solid circles) of photoluminescence obtained on a p-type InP sample immersed in superfluid helium (laser power $P \approx 200$ mW, excitation 11 900 cm^{-1}). The inset shows a typical photoluminescence spectrum obtained under low pumping level, and arrows give identification of observed lines according to Ref. 17.

plier followed by the current amplifier and two synchronous demodulators (lock-ins). Two types of experiments were performed. First, with low excitation power and high resolution, we looked for spectral features of the luminescence. Second with high excitation power (typically about 200 mW), and low resolution to measure polarization with a reasonable signal-to-noise ratio. This was necessary because of poor sensitivity of GaAs photomultiplier in the spectral range of interest. In polarization experiments the difference between σ^+ and σ^- polarization signals was measured by a lock-in amplifier fed with the reference from the photoelastic modulator. Photoluminescence intensity was measured by another lock-in amplifier fed with the reference from the chopper. The ratio of signals from both lock-ins gave directly the polarization degree of luminescence. Outputs of both lock-ins were digitized by a computer-controlled analog digital converter. Luminescence intensity and its degree of polarization were stored simultaneously.

Standard helium bath cryostat was used. Temperature was lowered down to 1.8 K by pumping of the helium bath and the samples were directly immersed in the superfluid helium. Magnetic fields parallel and perpendicular to the excitation beam were applied independently. A 2-T superconducting coil was used to apply a magnetic field parallel to the excitation beam. Another resistive coil placed outside the cryostat provided up to 300-G magnetic field perpendicular to the excitation beam. The degree of circular polarization was measured as a function of perpendicular (or parallel) magnetic field for different values of a constant parallel (perpendicular) magnetic field.

IV. RESULTS AND DISCUSSION

The inset of Fig. 1 shows a typical near-band-gap luminescence spectrum of a p-type InP sample at moderate excitation level. The spectrum is very similar to that reported by Fishman *et al.*[17] The three excitonic lines observed were attributed to a free exciton (X), an exciton bound to a neutral donor (D^0X), and an exciton bound to an ionized donor (D^+X). Photoluminescence studies on high-purity InP samples reveal more lines,[23] in particular those attributed to the (D^0X) complex which, according to Ref. 24, result from different angular momentum states of the $j=\frac{3}{2}$ hole. These lines are not resolved in doped samples. We note that in Ref. 23 the group of lines attributed to D^0X are situated around 1.417 eV rather than 1.415 eV, corresponding to the main peak in the inset in Fig. 1. Despite this slight difference we follow the identification of this line as (D^0X). The intensity and polarization spectra given in Fig. 1 were obtained under higher excitation level ($P \approx 200$ mW). The line attributed to D^+X has almost disappeared probably due to ionization of this weakly bound complex [activation energy of about 0.5 meV (Ref. 17)]. We estimate the density of photocarriers to be of the order of 10^{17} cm^{-3}, much higher than the neutral donor concentration (about 10^{15} cm^{-3}). The observed broadening of (D^0X) line is not surprising at this relatively high carrier density. It is even possible that at this high excitation level the luminescence is dominated by free excitons.

Note that a different assignment of the luminescence lines is possible. According to Ref. 25, the main luminescence line in Fig. 1 should be rather attributed to the exciton complex bound to acceptors. In the process of capture of a free exciton by an acceptor some depolarization should also occur. Its exact quantitative description is however not known. The interpretation of the structure seen in the polarization spectrum of Fig. 1 is beyond the scope of this paper, which is concerned with nuclear effects.

The Hanle depolarization curve measured at 11 411 cm^{-1} (excitation wave number 11 750 cm^{-1}) is Lorentzian with a half-width $B_H = 34$ G [see Fig. 2(a)]. Using Eqs. (7), (8), and (9), and the known value $g = 1.26$, we deduce $\tau \sim 9.3$ ns and $\tau_s \sim 2.7$ ns.

In configuration (b) with fixed circular polarization of the exciting laser beam we have looked for the manifestations of the nuclear field in the polarization of luminescence (mainly D^0X line). As explained in Sec. II, such effects can be observed if the magnetic field $\mathbf{B}$ is oblique with respect to the direction of excitation light. Indeed $\mathbf{B}_N$ is proportional to $(\mathbf{B}\cdot\mathbf{S})\mathbf{B}/B^2$, where $\mathbf{S}$ is the average electron-spin polarization. Hence $B_\parallel$ is necessary to have a nonzero B_N and $B_\perp$ allows its detection via the precession of $\mathbf{S}$. Two kinds of experiments have been done. (1) We sweep $B_\perp$ for a given $B_\parallel$. Typical polarization curves are shown in Fig. 2. (2) We sweep $B_\parallel$ for a given $B_\perp$. Typical polarization curves are shown in Fig. 3. In both cases the experimental curves are fitted with Eq. (12).

First of all, we note that the general appearance of the experimental dependencies of the degree of polarization on $B_\perp$ and $B_\parallel$ is in agreement with Eq. (12): the asymmetry in the $B_\parallel$ dependence and the additional maxima in the $B_\perp$ dependence clearly indicate the presence of the nuclear field. The additional maxima appear as a result of compensation of the external field by the nuclear magnetic field, as explained in Sec. II. This may happen if $\mathbf{B}\cdot\mathbf{S} < 0$, which corresponds to $z < 0$ [see Eqs. (5) and (12)]. Thus the positions of the additional maxima in Fig. 2 directly give the values of the nuclear magnetic field. The measured polarization values at these maxima are somewhat lower than $P(0)$, which may be quite plausibly explained by small spatial inhomogeneities of the nuclear field. Because of these inhomogeneities, the ex-

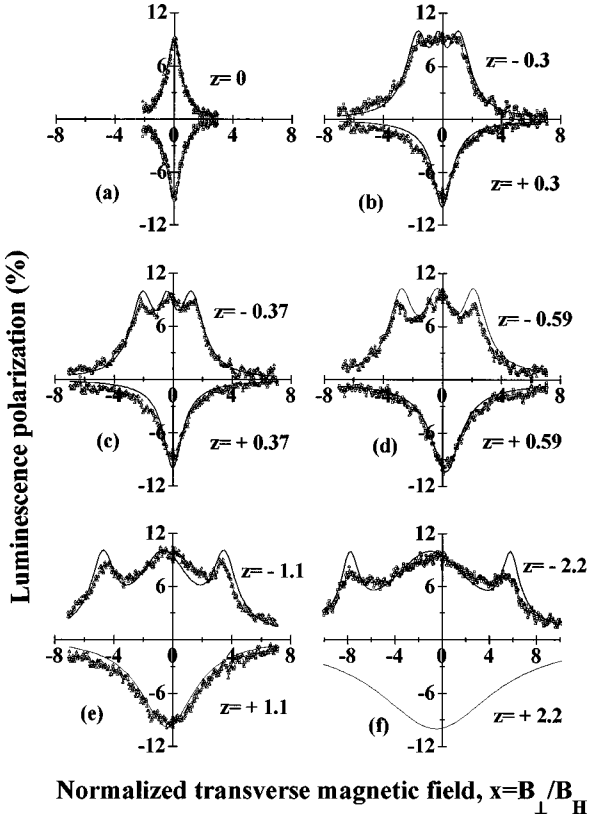

FIG. 2. Circular polarization degree of photoluminescence recorded at 11 410 cm^{-1}, vs the transverse magnetic field for different values of the dimensionless longitudinal field $z=B_{\parallel}/B_H$, $B_H=34$ G. (a) $z=0$, (b) $z=\pm 0.3$, (c) $z=\pm 0.37$, (d) $z=\pm 0.59$, (e) $z=\pm 1.1$, and (f) $z=\pm 2.2$. Full lines are fits by Eq. (12) with α depending on magnetic field, according to Fig. 4.

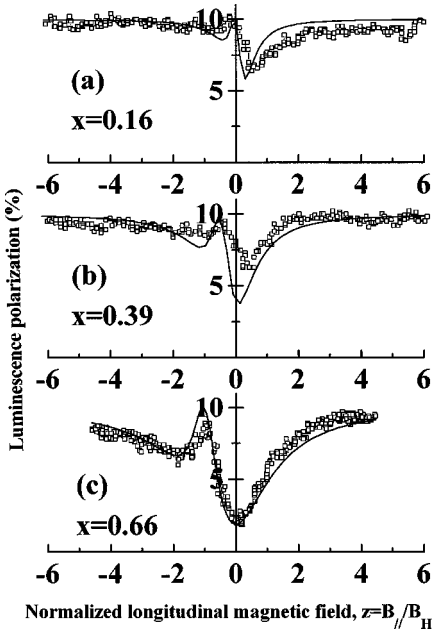

FIG. 3. Circular polarization degree of photoluminescence recorded at 11 410 cm^{-1}, vs the longitudinal magnetic field for different values of the dimensionless transverse field $x=B_{\perp}/B_H$, $B_H=34$ G. (a) $x=0.16$, (b) $x=0.39$, and (c) $x=0.66$. Full lines are fits by Eq. (12), with α depending on magnetic field, according to Fig. 4.

ternal field cannot be completely compensated for in the whole luminescence area. Each of the curves in Figs. 2 and 3 can be qualitatively (but not quantitatively) fit by Eq. (12), with a suitable choice of the parameter α. However, it is impossible to fit all of the data with a single α. We are thus led to the conclusion that this parameter is not a constant, but is a function of the magnetic field.

To find α as a function of the magnetic field, we proceed in the following way. We consider the set of $P(x)$ dependencies for different negative z, and measure the positions of the additional maxima x_m. Then we calculate $\alpha=(x_m^2+z^2)/|z|$, as follows from Eq. (14), and this value varies strongly for different z. We make the *assumption* that in fact α depends only on the absolute value of the magnetic field $(x^2+z^2)^{1/2}$, and accordingly we plot the obtained values of α against the variable $(x_m^2+z^2)^{1/2}$ (see Fig. 4). We fit the results by a second-order polynomial with three fitting parameters. It turns out that if we consider α in Eq. (12) as a function of $(x^2+z^2)^{1/2}$ obtained in such a manner, this equation now describes very well all of our experimental curves, not only for negative, but also for positive z (see the examples in Fig. 2). Also, the polarization measured as a function of z for different x is very well described without any additional fitting parameters (see the examples in Fig. 3). It should be noted that taking account of the magnetic-field dependence of α also improves the accuracy of the fits, compared to the case when the fits are done with $\alpha=$ const.

Let us discuss the possible origin of this strong magnetic-field dependence of α. All of the quantities entering Eq. (13), except f, are either constants, or experimentally measured values $P(0)$ and B_H. Thus the strong magnetic-field dependence observed should be attributed to the leakage factor f. Figure 5 gives this dependence derived from Eq. (13) and Fig. 4.

We see that in the available magnetic-field range the leakage factor increases strongly with magnetic field. Its maximal value, however, does not exceed 0.055, so that the measured nuclear magnetic field is always much smaller than

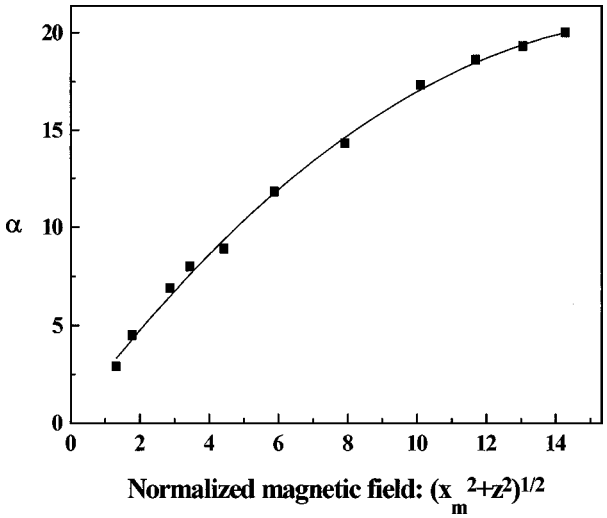

FIG. 4. Parameter α calculated from Eq. (14) vs $(x_m^2+z^2)^{1/2}$. The full line is a fit by a second-order polynomial with three fitting parameters.

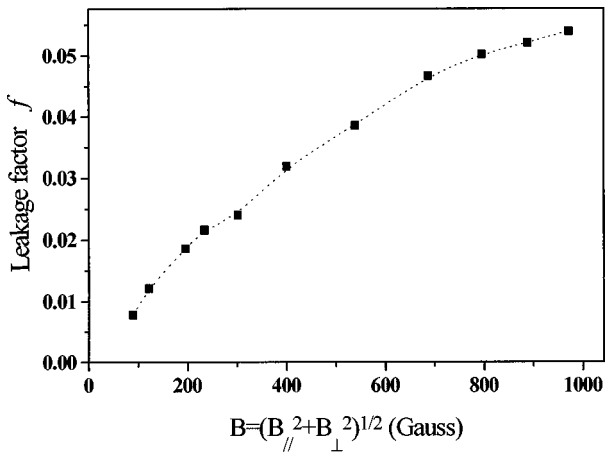

FIG. 5. Leakage factor f calculated from Eq. (13) and Fig. 4 vs the absolute value of the magnetic field. The line is a guide to the eye.

that expected on the basis of Eqs. (5) and (6). With this leakage factor the calculated critical temperature for self-polarization is $T_c \approx 0.3$ K.

The nature of the leakage is not clear. It does not seem probable that it is due to nuclear-spin-lattice relaxation processes, other than the hyperfine interaction with photocreated electrons, since at low temperatures such processes are normally rather slow (in GaAs, the dark nuclear relaxation times are on a scale of tens of seconds or even longer). While we especially looked for transient processes in the polarization of the luminescence, we did not observe any with a time scale longer than 1 s—the response time of our detection system, indicating fast electron-nuclear-spin exchange.

We think that the leakage may be understood qualitatively if we consider that the nuclei are subject to interaction not just with one group of spin-polarized photoexcited electrons, as was assumed in the derivation of Eq. (5), but with two or more electron species with different spin orientation and different relaxation times for the nuclei. In other words, there is a certain steady-state distribution of the photocreated electrons in energy, spin polarization, and location in space. Indeed, the polarization spectra in Fig. 1 demonstrate that a large portion of electrons responsible for the low-energy wing of the luminescence is not polarized. These electrons tend to depolarize the nuclei, and the resultant nuclear polarization should depend on the relative rates of nuclear relaxation by the polarized and nonpolarized electrons. The relaxation rate is proportional to the electron correlation time τ_c (the time that an electron spends within the interaction radius for a given nucleus). This in turn depends strongly on the degree of electron localization by donors and spatial fluctuations of the impurity potential. Since low-energy electrons are generally better localized, we could expect that the correlation times for nonpolarized electrons are longer than for the polarized ones. Hence the rate of nuclear relaxation by the nonpolarized electrons should be greater than the rate of the dynamic nuclear polarization by the polarized electrons. This could explain why the observed nuclear polarization is rather small.

This idea also helps us to understand qualitatively the increase of nuclear polarization with magnetic field. The relaxation time usually increases with magnetic field as $[1+(\omega\tau_c)^2]$, where ω is the electron Larmor precession frequency,[19] so that the longer τ_c is, the earlier the relaxation rate suppressed by the magnetic field. Thus the nuclear relaxation by the more localized, nonpolarized electrons, with longer τ_c, will be suppressed by lower magnetic field, than is the case for the polarized electrons, which should shift the balance toward an increase of nuclear polarization.

V. CONCLUSION

In summary, we have observed and measured optically induced nuclear magnetic fields in InP, using as a tool the oblique Hanle effect. Although the optical manifestations of the nuclear field are quite spectacular, and are well understood on the basis of a simple theory, the measured values are about 20 times smaller than one could expect under optimal conditions. Furthermore, we observed an unexpected strong dependence of the nuclear polarization on the applied magnetic field. In a qualitative manner, we explain these results as due to existence of several species of photocreated electrons with different degrees of localization and different correlation and relaxation times for the nuclei.

More detailed experimental studies of differently doped samples, with lower excitation power levels, and higher magnetic fields are needed to clarify the situation. It would be also interesting to investigate the nuclear effects on different parts of the polarization spectrum, to measure the contribution of nuclei located at different spatial regions of the crystal.

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*Permanent address: A. F. Ioffe Physico-Technical Institute, St. Petersburg, Russia.

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