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Low field electron-nuclear spin coupling in gallium arsenide under optical pumping conditions

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In a semiconductor, absorption of circularly polarized light (optical pumping) leads to spin-oriented photoelectrons. In this situation, the nuclei of the crystal are dynamically polarized through their hyperfine interaction with the electronic spins. Consequently the electrons experience a hyperfine magnetic field due to the oriented nuclei which may reach several kilogauss. When this large nuclear field is driven obliquely with respect to the direction of the exciting light, the precession of the electronic spins around the nuclear field leads to a decrease of the electronic polarization along the light excitation: it is a nuclear Hanle effect. This work is an experimental and theoretical study of these effects in weak external magnetic fields, of the order of the local field which characterizes the nuclear spin-spin interactions (a few gauss). Large nuclear fields are obtained at 77°K in strongly doped and compensated *p*-type GaAs samples. We present a model which includes the different effects of the hyperfine coupling when there is a nuclear spin temperature among all the nuclei of the sample: Dynamic polarization, nuclear field, but also, existence of an electronic field acting on the nuclei. We show that a small external field is able to drive the large nuclear field acting on the electrons; consequently the electronic polarization is very sensitive to external fields which are too small to have a direct effect on the electronic spin motion. We study experimentally the variation of the electronic polarization with the direction and magnitude of a small external magnetic field, by measuring the circular polarization of the luminescence light. The experimental results are in quantitative agreement with the theoretical predictions. The usual Hanle Lorentzian depolarization curve is strongly modified in low fields and *W*-like singularities appear around zero field. The experimental values of the average electronic and nuclear fields are in reasonable agreement with theoretical evaluations. These nuclear effects may strongly alter the measurement of the Hanle linewidth in standard optical pumping experiments.

I. INTRODUCTION

Interband optical pumping in semiconductors by absorption of circularly polarized light through the gap can lead to polarized photoelectrons. In the first experiment in silicon,[1] the Overhauser dynamic polarization of the nuclear spins of the crystal was used as a probe of the electronic polarization. In subsequent studies, the electronic polarization was measured from the degree of circular polarization of the luminescence light and the effect of the nuclei was not considered.[2,3] More recently[4-6] it has been shown that in several cases, due to the hyperfine interaction, the polarized nuclei create an internal magnetic field $\vec{B}_n$, acting on the electronic spins, which can be as high as several kilogauss. The effect of such a large field, which adds vectorially to the external field, can be observed when the electronic polarization is sensitive to the total field. This has permitted for instance the optical detection of nuclear magnetic resonance.[4,5,7] Furthermore, the oblique Hanle effect, that is the application of a static magnetic field oblique with respect to the incident light, has yielded a direct measurement of the nuclear field $\vec{B}_n$.[8] On the other hand, a wealth of unexplained effects in low magnetic fields (of the order of a few gauss) has been reported: The Hanle curve no longer has the classical Lorentzian shape but it is narrowed[9] or exhibits W-like shapes.[10] In some conditions, there appears hysteretic behavior[7] or oscillations in time of the electronic polarization.[11] All these effects are thought to be of nuclear origin. Let us also mention an experimental study of the nuclear effects when the external field and the exciting light polarization are modulated at the same frequency.[12]

In this paper, we present a theoretical and experimental study of low-field nuclear effects in doped gallium arsenide. We show that very small oblique fields (0.5 G) can modify the electronic polarization. To understand the system, it is necessary to consider simultaneously the existence of: (i) very large nuclear fields, (ii) nuclear spin temperature, (iii) electronic hyperfine magnetic fields acting on the nuclei, and (iv) feedback action between electronic and nuclear spins.

The reason why the electrons are extremely sensitive to small oblique external fields is that these small fields, together with the electronic field, assign the direction of the large internal nuclear field. This is the consequence of the rapid establishment of an internal equilibrium among the nuclear spins characterized by a spin temperature.[7,13] The nuclear field is then oblique with respect to light excitation, and causes Hanle depolarization of the electrons. The decrease of the electronic polarization in turn modifies the nuclear field causing a feedback action of the electrons on the nuclei.

We have been able to achieve quantitative agreement between the observed singularities of the Hanle effect and a theoretical model which combines the ideas mentioned above. This yields experimental determinations of the nuclear and electronic fields. We show theoretically and experimentally that *W*-like lines also appear in longitudinal fields whenever there exists a small transverse field. This study points out the possible importance in optical-pumping experiments of small parasitic fields such as the earth field.

An experimentally favorable situation is achieved when the hyperfine interaction is large, which leads to strong nuclear fields. This is the case when the electrons are localized. We work with *p*-type compensated GaAs samples ($N_A \sim 2\times 10^{18}$ cm^{-3}; $N_D/N_A \sim 0.3$) in which electrons are localized in potential wells, and where it has previously been shown that large nuclear polarizations occur under optical pumping conditions at liquid-nitrogen temperature.[8]

In Sec. II, we examine the relevant aspects of the hyperfine interaction. The theory is given in Sec. III, where the coupled equations governing the motion of the nuclear and electronic spins are derived. In Sec. IV, we present the experiments results which are discussed in Sec. V.

II. HYPERFINE COUPLING BETWEEN ELECTRONS AND NUCLEI

To understand the nature of the described effects, we first discuss the consequences of the hyperfine coupling between electronic and nuclear spins. The interaction of a single *s* electron of spin $\vec{S}$ with a nucleus of spin $\vec{I}$ is given by the Fermi contact Hamiltonian[14]:

$$\mathcal{H}_F = \frac{8\pi}{3}\frac{\mu_0}{4\pi} g_0 \mu_B \gamma_n \hbar \vec{I}\cdot\vec{S}\,\delta(\vec{r}), \tag{2.1}$$

where $\vec{r}$ is the coordinate of the electron relative to the nucleus, μ_B is the Bohr magneton taken as positive, g_0 the free-electron g factor,[15] and γ_n the nuclear-gyromagnetic ratio. Taking the average over the orbital coordinates of the electron, we obtain a spin Hamiltonian

$$\mathcal{H}'_F = \tfrac{2}{3}\mu_0 g_0 \mu_B \gamma_n \hbar \vec{I}\cdot\vec{S}\,|\psi(0)|^2, \tag{2.2}$$

where $|\psi(0)|^2$ is the value of the electronic wave function $\psi(\vec{r})$ at the position of the nucleus. The existence of this interaction has several well-known consequences in nuclear magnetism: Dynamic effects such as Korringa relaxation and dynamic polarization (Overhauser effect) and static effects such as the Knight shift of the nuclear resonance.[16] There are also the symmetrical effects of the nuclei on the electrons, namely the Overhauser shift of the conduction electron spin resonance,[17,18] and the effects of nuclear field on transport[19] and optical pumping.[4-12]

A. Dynamic effects of the hyperfine coupling

Dynamic nuclear polarization can appear when a system of nuclei relaxes at least partially, through the modulation of their hyperfine interaction with electronic spins which are out of thermal equilibrium. This is the case in optical pumping conditions. It can be shown that in the case where the only relaxation mechanism is the fluctuating part of the hyperfine interaction the nuclear polarization is[20]:

$$\langle\vec{I}\rangle = \frac{I(I+1)}{S(S+1)}\langle\vec{S}\rangle, \tag{2.3}$$

where $\langle\vec{S}\rangle$ is the static electronic spin.

Here, the electronic and nuclear thermal polarizations have been neglected, which is a correct approximation in low-field experiments. If one includes the other causes of nuclear relaxation (relaxation by unpolarized holes,[21] paramagnetic impurities,[22] quadrupolar relaxation[23]) the nuclear polarization is decreased by a leakage factor[16]

$$f = T_{1f}/(T_{1e}+T_{1f})\,; \tag{2.4}$$

T_{1e} and T_{1f} being, respectively, the relaxation times due to the coupling with the electrons and due to the other mechanisms. Let us mention that in principle there also exists a dynamic polarization of the electrons by the nuclei but this is very ineffective compared with the other mechanisms of electron spin relaxation.

B. Static effects

The interaction of one nucleus in the crystal with the electrons is obtained by summing the spin Hamiltonian defined in Eq. (2.2) over the occupied electronic states q. For a nucleus i at position $\vec{r}_i$ this interaction is equivalent to a Zeeman interaction in a magnetic field $\vec{B}_e^i$ of electronic origin:

$$\vec{B}_e^i = -\frac{2\mu_0}{3} g_0 \mu_B \sum_q \vec{S}_q |\psi_q(\vec{r}_i)|^2 . \tag{2.5}$$

In the same way, the spin of an electron in an orbital state ψ_q of effective g-factor g^* feels an internal field of nuclear origin which is equivalent to an external field of value

$$\vec{B}_n^q = \frac{2\mu_0}{3}\frac{g_0}{g^*}\hbar \sum_i \gamma_i \vec{I}_i |\psi_q(r_i)|^2 , \tag{2.6}$$

where g^* is the effective g factor of the electron.

We now examine the characteristics of the coupling in the case of gallium arsenide under conditions of optical pumping, i.e., we discuss the nature of the electronic states and of the nuclei involved and we give an order of magnitude of the corresponding electronic and nuclear fields. There are three types of nuclei, all of spin $\frac{3}{2}$: ^{69}Ga, ^{71}Ga, and ^{75}As. (In the following the nuclear species are referred to by the subscript α.) The electrons occupy different types of orbital states, either localized or delocalized. They can also form excitons or excitonic complexes but these last cases are more complicated due to the hyperfine interaction of the holes with the nuclei and will not be examined here. The localized states can be of different nature according to the samples used: In pure samples the electrons can be trapped on donors.[7] The case of strongly doped samples is more complicated due to the presence of more than one impurity within an electronic Bohr radius. Although the experiments were performed in the latter case we shall give an evaluation of the parameters of the coupling in the case of electrons trapped on donors which is simpler and constitutes a simplified picture for our case. Since we are considering states near the bottom of the conduction band, we write the orbital function of the qth electron as a product of an envelope wave function $F_q(r)$ by the rapidly varying periodic function $u_0'(\vec{r})$

$$\psi_q(\vec{r}) = F_q(\vec{r})u_0'(\vec{r}) = F_q(\vec{r})u_0(r)(V/\Omega)^{1/2} , \tag{2.7}$$

where $u_0(r)$ is the $\vec{k}=0$ Bloch state, V is the volume of the sample, and Ω the volume of the unit cell. The function $u_0'(r)$ is the conveniently renormalized function which is directly comparable to the wave function of the atom. This function takes a different value on cation and anion sites. The values of the electronic density $d_\alpha = |u_0'(r_\alpha)|^2$ on the nuclei α are evaluated in Appendix A. These values are

$$\begin{aligned} d_{(^{75}\mathrm{As})} &\simeq 9.8\times 10^{25}\ \mathrm{cm}^{-3} , \\ d_{(^{69}\mathrm{Ga})} = d_{(^{71}\mathrm{Ga})} &\simeq 5.8\times 10^{25}\ \mathrm{cm}^{-3} , \end{aligned} \tag{2.8}$$

The localization or delocalization of the electrons is described by the envelope part $F_q(r)$ of the wave function. In the case of an electron at the bottom of the conduction band, the value of $F_q(\vec{r})$ is $(\Omega/V)^{1/2}$ inside the crystal and zero outside. In the case of a donor state we have[24]:

$$F_q(\vec{r}) = (\Omega/\pi a_0^{*3})^{1/2}\exp(-r/a_0^*) , \tag{2.9}$$

where a_0^* is the effective radius of the shallow donor state ($a_0^* \simeq 100$ Å in GaAs). The norm of these envelope functions is

$$\int |F_q(r)|^2 d^3r = \Omega . \tag{2.10}$$

It is now possible to calculate the electronic field seen by a nucleus due to free or localized electrons. For free electrons this field which is independent of the position of the nucleus is very small. For example, it has been measured to be 1 G for 10^{15} totally polarized electrons in InSb, where hyperfine interactions are strong[25] and will be neglected here. For localized electrons the only nuclei which experience a nonvanishing hyperfine interaction are those situated in the vicinity of an occupied center. They feel the electronic field of only one electron at a time, the electron trapped on the donor. In this case, the electronic field $B_e^\alpha(r)$ seen by a nucleus α at distance r from the donor is obtained from Eqs. (2.5), (2.8), and (2.9):

$$\vec{B}_e^\alpha(r) = b_e^\alpha(r)\vec{S} = b_e^\alpha(0)\vec{S}\exp(-2r/a_0^*) \tag{2.11}$$

with

$$b_e^\alpha(0) = -\tfrac{2}{3}\mu_0 g_0 \mu_B(\Omega/\pi a_0^{*3})d_\alpha . \tag{2.12}$$

The numerical estimate of $b_e^\alpha(0)$ for the three nuclear species gives

$$\begin{aligned} b_e^{(^{75}\mathrm{As})}(0) &\simeq -220\ \mathrm{G} , \\ b_e^{(^{69}\mathrm{Ga})}(0) = b_e^{(^{71}\mathrm{Ga})}(0) &\simeq -130\ \mathrm{G}. \end{aligned} \tag{2.13}$$

The static part of this electronic field is given by Eq. (2.11) where the instantaneous electronic spin $\vec{S}$ is replaced by its static average $\langle\vec{S}\rangle$. If the donor is occupied a fraction Γ_t of the total time the static electronic field $\langle\vec{B}_e^\alpha(r)\rangle$ is reduced

$$\langle\vec{B}_e^\alpha(\vec{r})\rangle = \Gamma_t b_e^\alpha(0)\langle\vec{S}\rangle\exp(-2r/a_0^*) . \tag{2.14}$$

It is the fluctuating part of the electronic field which is responsible for the dynamic polarization of the nuclei (the relaxation by conduction electrons is negligible in our case[21]). An order of magnitude of the polarization time of the nuclei α at distance r from an impurity is given by[26]

$$\frac{1}{T_{1e}^\alpha(r)} \simeq \Gamma_t[\gamma_\alpha b_e^\alpha(0)]^2\tau_c\exp(-4r/a_0^*) , \tag{2.15}$$

where τ_c is the correlation time of the relaxing interaction. Here, we can take τ_c of the same order of magnitude as the lifetime τ of the trapped electron because it is the capture and recombination of the electrons which modulate the hyperfine interaction. One finds $T^{\alpha}_{1e}(0)$ in the range 10^{-1}–10^{-2} sec with τ_c ranging between 10^{-8} and 10^{-9} sec and Γ_t equal to unity. The relaxation time of the total system of nuclear spins may be influenced by spin diffusion[27] but when the interimpurity distance is small this should not play a significant role. This is the case of our highly doped samples and a value of the nuclear relaxation time can be estimated from Eq. (2.15) with $r \simeq a_0^*$.

The nuclear magnetic field experienced by an electron in an orbital state ψ_q is the sum of the contributions $\vec{B}^q_\alpha$ of the three nuclear species, which are given by equations of the form of Eq. (2.6) where the sum is restricted to the nuclei of species α of the crystal. If all the nuclei of the same species have the same polarization throughout the sample, the quantity $\langle \vec{I}_{i\in\alpha}\rangle = \langle \vec{I}_\alpha\rangle$ is independent of the position of the nucleus i and can be factorized. The nuclear field $\vec{B}^q_\alpha$ due to the nuclear species α is given by

$$\vec{B}^q_\alpha = \frac{2\mu_0}{3}\frac{g_0}{g^*}\hbar\gamma_\alpha\langle\vec{I}_\alpha\rangle\sum_{i\in\alpha}|\psi_q(\vec{r}_i)|^2 ,$$

$$\vec{B}^q_\alpha = \frac{2\mu_0}{3}\frac{g_0}{g^*}\hbar\gamma_\alpha d_\alpha\langle\vec{I}_\alpha\rangle\sum_{i\in\alpha}|F_q(\vec{r}_i)|^2 . \qquad (2.16)$$

Due to the slowly varying character of the envelope function $F_q(\vec{r})$ and using Eq. (2.10), one sees that the nuclear field created by a uniform nuclear polarization is independent of the electronic state,

$$\vec{B}_\alpha = \tfrac{2}{3}\mu_0(g_0/g^*)\hbar\gamma_\alpha x_\alpha d_\alpha\langle\vec{I}_\alpha\rangle = b_\alpha\langle\vec{I}_\alpha\rangle . \qquad (2.17)$$

Here, x_α is the concentration of the nuclear species

$$x_{(^{75}\mathrm{As})} = 1 , \quad x_{(^{69}\mathrm{Ga})} = 0.60 , \quad x_{(^{71}\mathrm{Ga})} = 0.40 . \qquad (2.18)$$

The numerical values of b_α are (using $g^* = -0.44$)[15]

$$b_{(^{75}\mathrm{As})} \simeq -18.4\ \mathrm{kG}, \qquad b_{(^{69}\mathrm{Ga})} \simeq -9.1\ \mathrm{kG},$$

and (2.19)

$$b_{(^{71}\mathrm{Ga})} \simeq -7.8\ \mathrm{kG} .$$

When the polarization of the nuclei is not uniform (because only the nuclei near donors are rapidly polarized) a conduction electron sees a weak nuclear field because only a small fraction of the nuclear spins I_i contributes to the sum of Eq. (2.6). However, in this same case the electron localized on a donor sees a strong nuclear field because in Eq. (2.6) the values of $|\psi_q(r_i)|^2$ near the donor are large due to the localization. If one supposes that the nuclear polarization is uniform near the donor center up to a distance of the order of a few Bohr radii Eq. (2.6) gives the same value for the nuclear field as in the case of completely uniform polarization. The nuclear field is again given by Eq. (2.17). Taking together Eqs. (2.3), (2.17), and (2.19) we see that with a mean electronic spin $\langle S\rangle \simeq 0.1$ one expects nuclear fields of the order of a few teslas if there is no leak. These giant nuclear fields can produce spectacular effects on optical pumping as we show in Sec. III.

III. THEORY

In this section, we obtain the equations governing the motion of the electronic magnetization under conditions of optical pumping, including the effects of the hyperfine coupling. Our experiments are made under steady-state regime: the characteristic time of the experiment is much larger than the three times which are relevant to our system: The electronic relaxation time (10^{-9} sec), the nuclear spin-spin relaxation time ($\simeq 10^{-4}$ sec), and the nuclear spin-lattice relaxation time ($\simeq 10^{-1}$ sec). For this reason we calculate the steady-state value of the electronic spin in a given total static field which is the sum of the external field and of the steady-state nuclear field. The nuclear field itself is obtained by calculating the steady-state nuclear magnetization due to dynamic polarization, taking correctly into account the nuclear spin-spin interactions.

As we have seen, the physical system under consideration is very complex: (i) There are three types of nuclei; (ii) gallium and arsenic see different electronic fields, which may vary with the position of the nucleus; (iii) the nuclei under consideration have quadrupolar moments and although the quadrupolar interaction is zero in the zinc-blende structure there could be quadrupolar effects related to impurities[28-30]; (iv) the electrons responsible for the dynamic polarization are localized in potential wells due to the random distribution of donors and acceptors so that the electronic wave functions are not well known. Furthermore in our strongly doped ($N_A \simeq 2\times10^{18}\ \mathrm{cm}^{-3}$) and compensated samples the interimpurity distance is of the same order as the effective Bohr radius and the spin diffusion characteristic lengths.[27] Consequently we do not know to what extent the nuclear polarization is homogeneous.

A microscopic description of the nuclear relaxation in this case is beyond the scope of this paper. To deal with such a situation we shall consider in the theory the following simplified picture: All the electrons are in the same orbital states. This means that we neglect the dispersion in the characteristics of the localization centers. We also suppose that the nuclei of interest (near localization centers) experience the same static electronic field, which we write as

$$\vec{B}_e^\alpha = \Gamma_s \langle \vec{B}_e^\alpha(0)\rangle = \Gamma_s \Gamma_t b_e^\alpha(0)\langle \vec{S}\rangle = \Gamma b_e^\alpha(0)\langle \vec{S}\rangle . \tag{3.1}$$

The factor Γ_s which is smaller than unity expresses that this average electronic field is smaller than the maximum static electronic field $\Gamma_t b_e^\alpha(0)\langle\vec{S}\rangle$ defined in Eq. (2.14).

In first approximation, we neglect the possible quadrupolar couplings. We suppose that all the nuclei α of interest have the same polarization $\langle I_\alpha\rangle$ so that they create a nuclear field given by Eq. (2.17). The total nuclear field $\vec{B}_n$ seen by an electron is then

$$\vec{B}_n = \sum_\alpha \vec{B}_\alpha = \sum_\alpha b_\alpha \langle \vec{I}_\alpha\rangle . \tag{3.2}$$

In this picture, all the electrons see the same nuclear field $\vec{B}_n$ and it is sufficient to study the motion of an individual electronic spin.

A. Motion of the electronic spin

We can write the evolution of the electronic spin $\langle\vec{S}\rangle$ as

$$\frac{d\langle\vec{S}\rangle}{dt} = \frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{creat.}} + \frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{rec.}} + \frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{relax.}} + \frac{\partial\langle S\rangle}{\partial t}\bigg|_{\text{field}} + \frac{\partial\langle S\rangle}{\partial t}\bigg|_{\text{nucl.}} . \tag{3.3}$$

The first term $(\partial\langle\vec{S}\rangle/\partial t)_{\text{creat.}}$ expresses the transfer of angular momentum from exciting light to electrons. The second and third terms account for the evolution of the mean electronic spin due to recombination and spin relaxation of the photoexcited electrons. The three first terms can be written[1-3]

$$\frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{creat.}} + \frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{rec.}} + \frac{\partial\langle\vec{S}\rangle}{\partial t}\bigg|_{\text{relax.}} = -\frac{\langle\vec{S}\rangle - \langle\vec{S}_0\rangle}{T_{1*}^e} . \tag{3.4}$$

Here, the total electronic relaxation rate is

$$1/T_{1*}^e = 1/T_1^e + 1/\tau , \tag{3.5}$$

where τ is the recombination time and T_1^e the electronic relaxation time. The steady-state spin $\langle\vec{S}_0\rangle$ is directed along the direction of propagation of the light. In III–V compounds and for σ^+ light of energy close to the band gap, the value of $\langle S_0\rangle$ is[1-3]

$$\langle S_0\rangle = -\tfrac{1}{4} T_1^e/(T_1^e + \tau) . \tag{3.6}$$

If one adds the term which describes the Larmor precession of the electronic spin in the external magnetic field $\vec{B}$ one obtains a Bloch-like equation

$$\frac{d\langle\vec{S}\rangle}{dt} = -\frac{\langle\vec{S}\rangle - \langle\vec{S}_0\rangle}{T_{1*}^e} - \frac{g^*\mu_B}{\hbar}\langle\vec{S}\rangle\times\vec{B} . \tag{3.7}$$

This equation holds provided the electronic relaxation times T_1^e and T_2^e of the components of the electronic spin parallel and perpendicular to the magnetic field are the same. This is true in the case of rapid motion known as the "extreme narrowing case".[31]

In the following we always choose $\vec{B}$ in the xz plane, z being the direction of propagation of the incident light. In a purely transverse external field ($B_z = B_y = 0$), the steady-state solution of Eq. (3.7) gives for $\langle S_z\rangle$ the usual Lorentzian decrease characteristic of the Hanle effect

$$\langle S_z\rangle = \langle S_0\rangle \Delta B^2/(\Delta B^2 + B_x^2) \tag{3.8}$$

with a half-width

$$\Delta B = \hbar(|g^*|\mu_B T_{1*}^e)^{-1} . \tag{3.9}$$

Equations (3.6) and (3.8) are the usual equations of optical pumping when nuclear effects are absent.

The possible actions of the nuclei have been described in Sec. II. We recall that the nuclei do not dynamically polarize the electrons. Their only effect is to create a static nuclear field $\vec{B}_n$ the existence of which has two consequences; firstly, the electronic spin precesses around the nuclear field according to

$$\left(\frac{\partial\langle\vec{S}\rangle}{\partial t}\right)_{\text{nucl.}} = -\frac{g^*\mu_B}{\hbar}\langle\vec{S}\rangle\times\vec{B}_n . \tag{3.10}$$

Secondly, $\vec{B}_n$ can modify the electronic spin relaxation time[5] and consequently the value of $\langle S_0\rangle$. In our experimental case, this last effect is weak and will be neglected in first approximation. Finally, the total evolution of the electronic spin can be written from Eqs. (3.7) and (3.10)

$$\frac{d\langle\vec{S}\rangle}{dt} = -\frac{\langle\vec{S}\rangle - \langle\vec{S}_0\rangle}{T_{1*}^e} - \frac{g^*\mu_B}{\hbar}\langle\vec{S}\rangle\times(\vec{B}+\vec{B}_n) . \tag{3.11}$$

This is the fundamental equation of the system. The nuclear field is calculated in the following subsection: It is shown that $\vec{B}_n$ is of the form

$$\vec{B}_n = \vec{B}_n' + A\langle\vec{S}\rangle . \tag{3.12}$$

The term $A\langle\vec{S}\rangle$ plays no role in Eq. (3.11) so that the steady-state equation can be written

$$(g^*/|g^*|)\Delta B(\langle\vec{S}\rangle - \langle\vec{S}_0\rangle) + \langle\vec{S}\rangle\times(\vec{B}+\vec{B}_n') = 0 , \tag{3.13}$$

where $\vec{B}_n'$ plays the role of an "effective nuclear field."

B. Calculation of the nuclear field

In the small external fields where we work, the Zeeman interaction of one given nuclear spin in the solid is not greater than the magnetic interactions of this nucleus with its neighbors. In this case, it is well-known that the effect of the nuclear spin-spin interactions is the establishment of a thermal equilibrium among the nuclear spins in a time of the order of the inverse width of the NMR line, ~100 μsec. The nuclear system is then characterized by a spin temperature[32] θ (of course this is true only if the driving forces on the system, light and magnetic field, can be considered as static during this time interval[33,34]). It is no longer possible to speak of one individual nuclear spin, and the density matrix which describes the total nuclear system is of the form (in the high-temperature approximation)

$$\sigma = (\underline{I} - \mathcal{H}_0/k_B\theta)(\mathrm{Tr}\,\underline{I})^{-1} . \tag{3.14}$$

Quite generally, the static Hamiltonian $\mathcal{H}_0$ is the sum of the Zeeman energies Z_i of all the spins in the external field $\vec{\mathrm{B}}$ and in the static part $\langle\vec{\mathrm{B}}_e^i\rangle$ of the hyperfine field $\vec{\mathrm{B}}_e^i$ defined by Eq. (2.5), of the spin-spin interaction $\mathcal{H}_{ss}$ and of the possible quadrupolar couplings which we neglect. The expression of $\mathcal{H}_0$ is given by

$$\mathcal{H}_0 = \sum_i Z_i + \mathcal{H}_{ss} = -\sum_i \gamma_i \hbar(\vec{\mathrm{B}} + \langle\vec{\mathrm{B}}_e^i\rangle)\cdot\vec{\mathrm{I}} + \mathcal{H}_{ss} . \tag{3.15}$$

The spin-spin interaction $\mathcal{H}_{ss}$ is the sum over all spins of dipolar, pseudodipolar and exchange interactions.[35,36]

$$\mathcal{H}_{ss} = \sum_{i>j} (\mathcal{H}_{D,ij} + \mathcal{H}_{PD,ij} + \mathcal{H}_{E,ij}) . \tag{3.16}$$

The expression of the dipolar interaction is

$$\mathcal{H}_{D,ij} = \sum_{i>j} \frac{\gamma_i\gamma_j\hbar^2}{r_{ij}^3}\left(\vec{\mathrm{I}}_i\cdot\vec{\mathrm{I}}_j - \frac{3(\vec{\mathrm{I}}_i\cdot\vec{\mathrm{r}}_{ij})(\vec{\mathrm{I}}_j\cdot\vec{\mathrm{r}}_{ij})}{r_{ij}^2}\right), \tag{3.17}$$

where $\vec{\mathrm{r}}_{ij}$ is the vector joining i and j. The pseudodipolar and exchange interactions are

$$\mathcal{H}_{PD,ij} = \mathcal{B}_{ij}\mathcal{H}_{D,ij} , \tag{3.18}$$

$$\mathcal{H}_{E,ij} = a_{ij}(\gamma_i\gamma_j\hbar^2/r_{ij}^4)\vec{\mathrm{I}}_i\cdot\vec{\mathrm{I}}_j , \tag{3.19}$$

where $\mathcal{B}_{ij}$ and a_{ij} are scalar quantities.

The fact that $\langle\vec{\mathrm{B}}_e^i\rangle$ depends on nuclear species and position does not preclude the establishment of a unique spin temperature as long as the difference between the Zeeman energies of two neighboring spins is not larger than the spin-spin energies. This is the case here. As said before, we consider for simplicity the case where all the nuclei α experience the same electronic field which is given by Eq. (3.1). The total field seen by a nucleus of species α is

$$\vec{\mathrm{B}}_T^\alpha = \vec{\mathrm{B}} + \Gamma b_e^\alpha(0)\langle\vec{\mathrm{S}}\rangle . \tag{3.20}$$

The static Hamiltonian $\mathcal{H}_0$ becomes

$$\mathcal{H}_0 = \sum_\alpha Z_\alpha + \mathcal{H}_{ss} \tag{3.21}$$

with

$$Z_\alpha = -\gamma_\alpha\hbar\vec{\mathrm{B}}_T^\alpha\cdot\sum_{i\in\alpha}\vec{\mathrm{I}}_i . \tag{3.22}$$

The expectation value of any operator is then obtained by the standard formula

$$\langle A\rangle = \mathrm{Tr}\,\sigma A . \tag{3.23}$$

This gives

$$\langle\vec{\mathrm{I}}_\alpha\rangle = \hbar\gamma_\alpha\frac{I_\alpha(I_\alpha+1)}{3}\frac{1}{k_B\theta}[\vec{\mathrm{B}} + \Gamma b_e^\alpha(0)\langle\vec{\mathrm{S}}\rangle] . \tag{3.24}$$

$$\frac{\langle Z_\alpha\rangle}{c_\alpha(B_T^\alpha)^2} = \frac{\langle\mathcal{H}_{ss}\rangle}{B_L^2} = \langle\mathcal{H}_0\rangle\left(B_L^2 + \sum_\alpha c_\alpha(B_T^\alpha)^2\right)^{-1}$$

$$= -\frac{1}{k_B\theta}\sum_\alpha\frac{n_\alpha}{3}I_\alpha(I_\alpha+1)(\gamma_\alpha\hbar)^2 , \tag{3.25}$$

where n_α is the number of spins of species α and

$$c_\alpha = n_\alpha I_\alpha(I_\alpha+1)\gamma_\alpha^2\bigg/\left(\sum_\alpha n_\alpha I_\alpha(I_\alpha+1)\gamma_\alpha^2\right) . \tag{3.26}$$

The local field B_L is defined by

$$B_L^2 = \mathrm{Tr}\,\mathcal{H}_{ss}^2\bigg/\left(\tfrac{1}{3}\mathrm{Tr}\sum_\alpha n_\alpha\gamma_\alpha^2\hbar^2 I_\alpha^2\right) . \tag{3.27}$$

It is a measure of the strength of the spin-spin interactions. Detailed studies of NMR and nuclear acoustic resonance (NAR) have been performed in GaAs.[30,37] They yield the values of the different spin-spin interactions so that it is possible to compute B_L^2 for GaAs. This is done in Appendix B and gives

$$B_L^2 = 2.1 \pm 0.1\ \mathrm{G}^2 . \tag{3.28}$$

Equations (3.24) and (3.25) state that: (i) the nuclear magnetizations $n_\alpha\gamma_\alpha\hbar\langle\vec{\mathrm{I}}_\alpha\rangle$ are directed along the total field seen by the nuclei of species α; (ii) there is no nuclear magnetization when this field is zero; (iii) the partition of the internal energy $\langle\mathcal{H}_0\rangle$ between the Zeeman energies $\langle Z_\alpha\rangle$ and the spin-spin energy $\langle\mathcal{H}_{ss}\rangle$ depends on the relative values of the magnetic fields B_T^α and of the local field B_L.

The calculation of the steady-state nuclear magnetization must take two processes into account simultaneously: The thermalization of the nuclear spin system along the magnetic field that they see and the dynamic polarization by the spin-oriented electrons which tends to polarize equally all the nuclei parallel to the mean electronic spin

[Eq. (2.3)]. The values of the steady-state nuclear magnetizations are determined by the balance of the polarization and of the relaxation mechanisms of the Zeeman and spin-spin energies. We calculate the spin temperature in the general framework of the evolution of the nuclear density matrix $\underline{\sigma}$: The relaxing interaction $\mathcal{H}'(t)$ is the sum of $\underline{\text{the}}$ interaction with the electrons $\mathcal{H}'_e(t)$ which tends to polarize the nuclei and of the other relaxing mechanisms $\mathcal{H}'_f(t)$ which have a depolarizing effect (leak):

$$\mathcal{H}'(t)=\mathcal{H}'_e(t)+\mathcal{H}'_f(t)=\sum_\mu \mathcal{H}'_\mu(t)\,, \tag{3.29}$$

where μ is e or f.

The time evolution of the density matrix σ is in the extreme narrowing case[31,32]

$$\frac{d\sigma}{dt}=-i[\mathcal{H}_0,\sigma] - \sum_\mu \int_0^\infty d\tau\langle[\mathcal{H}'_\mu(t),[\mathcal{H}'_\mu(t-\tau),\sigma-\sigma_\mu]]\rangle\,. \tag{3.30}$$

The angular brackets symbolize an average over a statistical ensemble. This equation expresses that the system tends to evolute towards two different states σ_μ under the influences of the polarized electrons (σ_e) and of the other relaxing interactions (σ_f). It is natural to assume that σ_e is diagonal in the representation which diagonalizes the component of the nuclear spins along the electronic spins $\langle\vec{S}\rangle$ and has the following form which gives the value (2.3) for the dynamic nuclear polarization:

$$\sigma_e=\left(\underline{I}+\sum_\alpha \frac{3}{S(S+1)}\sum_{i\in\alpha}\vec{I}_i\cdot\langle\vec{S}\rangle\right)\Big/\mathrm{Tr}\,\underline{I}\,. \tag{3.31}$$

The density matrix σ_f is simply proportional to the unity matrix:

$$\sigma_f=\underline{I}/\mathrm{Tr}\,\underline{I}\,. \tag{3.32}$$

Because the magnetic fields and temperature used in our experiments are such that the usual thermal nuclear polarization is vanishingly small.

With Eqs. (3.14)–(3.20) and (3.30) it is possible to express that the total energy of the system is static. This yields the steady-state spin temperature, from which we obtain the nuclear magnetizations by Eq. (3.24), and finally, the nuclear fields acting on the electrons by Eq. (2.17). From Eq. (3.24) one sees that the nuclear field is the sum of two terms which are, respectively, along the external field and along the electronic field. We know from Eq. (2.12) and (2.13) that this last term can be dropped so that the effective nuclear field is of the form

$$\vec{B}'_n=K\vec{B}\,. \tag{3.33}$$

The calculation is somewhat complicated because there are three different types of nuclei which see different magnetic fields. It is done in Appendix B, where we consider for $\mathcal{H}'_\mu(t)$ the case of scalar interactions with complete correlation of the fluctuating fields seen by two neighboring nuclei. The final expresssions are cumbersome. They can be simplified if one supposes that the electronic field is smaller than the local field. In this case we find (B28)

$$\vec{B}'_n=f\,b_n\frac{\langle\vec{S}\rangle\cdot(\vec{B}+\Gamma b_e\langle\vec{S}\rangle)}{B^2+\xi B_L^2}\vec{B} \tag{3.34}$$

Here, f is an equivalent leakage factor [Eq. (2.4)] and b_n and b_e are suitable averages of the nuclear and electronic fields b_α and $b_e^\alpha(0)$ given by Eqs. (3.1), (2.13), and (2.19). A numerical evaluation discussed in Appendix B for the case of GaAs gives

$$b_n\simeq-170\text{ kG}\,, \tag{3.35}$$

$$b_e\simeq-170\text{ G}\,. \tag{3.36}$$

The quantity ξ is a measure of the relative relaxation rates of the spin-spin and Zeeman energies. When there is one spin species and no exchange interaction $\xi=3$ in the case of complete spatial correlation of the relaxing interactions. If exchange interactions are present ξ is reduced because the exchange energy between like spins is not relaxed by correlated interactions. In Appendix B we obtain $\xi\simeq2.2\pm0.3$ for GaAs and the theoretical spin-spin parameter ξB_L^2 is

$$\xi B_L^2=4.6\pm0.5\text{ G}^2\,. \tag{3.37}$$

Let us remark the simplicity of Eq. (3.34) which shows that, although the magnetizations of the different nuclear species are aligned along different directions, the whole system behaves as one unique "equivalent" nuclear-spin system characterized by the three quantities $f b_n$, Γb_e, and ξB_L^2.

C. Coupled system

The equations of the electron nuclear system in steady-state regime (3.13), (3.33), and (3.34) can be written

$$(g^*/|g^*|)\Delta B(\langle\vec{S}\rangle-\langle\vec{S}_0\rangle)+(1+K)\langle\vec{S}\rangle\times\vec{B}=0\,, \tag{3.38}$$

$$K=f\,b_n\frac{(\vec{B}+\Gamma b_e\langle\vec{S}\rangle)\cdot\langle\vec{S}\rangle}{B^2+\xi B_L^2}\,. \tag{3.39}$$

From Eq. (3.38), one obtains

$$(\vec{B}+\Gamma b_e\langle\vec{S}\rangle)\cdot\langle\vec{S}\rangle=\langle S_0\rangle(B_z+\Gamma b_e\langle S_z\rangle)\,, \tag{3.40}$$

and finally we write the coupled system of equations which permit us to calculate the electronic polarization for any given external field

$$\frac{\langle S_z\rangle}{\langle S_0\rangle}=\frac{\Delta B^2+B_z^2(1+K)^2}{\Delta B^2+B^2(1+K)^2}\,, \tag{3.41}$$

$$K=fb_n\langle S_0\rangle\frac{B_z+\Gamma b_e\langle S_z\rangle}{B^2+\xi B_L^2}\;. \tag{3.42}$$

Equation (3.41) is the usual equation of the oblique Hanle effect but here the external field is multiplied by an amplification factor K of nuclear origin. This amplification factor is of the order of $fb_n\Gamma b_e\langle S_0\rangle^2$. With $f=1$, $\Gamma=1$, $\langle S_0\rangle=\frac{1}{2}$ and the above values of b_n, b_e, and ξB_L^2 we obtain $K\sim 10^6$. Even with smaller polarizations and leakage factors, the factor K may be much larger than unity and, consequently, it is possible for small external fields to produce strong electronic depolarizations.

We point out that the value of K depends on the electronic polarization via the electronic field $\Gamma b_e\langle S_z\rangle$ and the behavior of the system can be influenced by the resulting feedback action of the nuclei on the electrons. The calculation of the electronic polarization from Eqs. (3.41) and (3.42) leads to a third-degree equation. When it has three real roots the actual state of the system is not uniquely determined. This may lead to irreversibilities when the external field is swept.[7] In the present work, we restrict ourselves to conditions where there is only one real solution, i.e., where the coupling is not too strong (small electronic fields).

We consider first the simplified situation where the electronic field is zero ($b_e=0$). In this case Eq. (3.41) describes the oblique Hanle effect in a total field $(1+K)\vec{B}$ which is independent of the electronic polarization $\langle S_z\rangle$. For the small values of the external field that we consider, ($B\ll\Delta B$) Eqs. (3.41) and (3.42) show that $\langle S_z\rangle$ is equal to $\langle S_0\rangle$ when the external field is purely longitudinal or purely transverse. Indeed, a longitudinal nuclear field has no effect besides its possible effect on the electronic relaxation time T_1^e.[5] On the other hand, in a transverse magnetic field the assigned direction of the nuclear magnetization is perpendicular to the electronic spin and the nuclei cannot get polarized. On the contrary, in an oblique external field there is a decrease of $\langle S_z\rangle$. Therefore, if we sweep through zero the longitudinal field in a given transverse field or if, conversely, we sweep the transverse field in a given longitudinal field, we should observe symmetrical W-shaped lines.

If we now take into account the existence of the small electronic field the overall picture of the system is not changed very much in a longitudinal sweep except that the center of the line is shifted ($\langle S_z\rangle=\langle S_0\rangle$ when $B_z=-\Gamma b_e\langle S_0\rangle$) whatever the magnitude of the transverse field: For this value of B_z the nuclei see a purely transverse total field along which they cannot get polarized. On the other hand if the transverse field is scanned, a W-shaped line will be observed *even when no external longitudinal field is applied*: Due to the existence of the electronic field the total field seen by the nuclei is oblique and the resulting nuclear field causes an electronic depolarization. The geometry of the system in this case is pictured on Fig. 1.

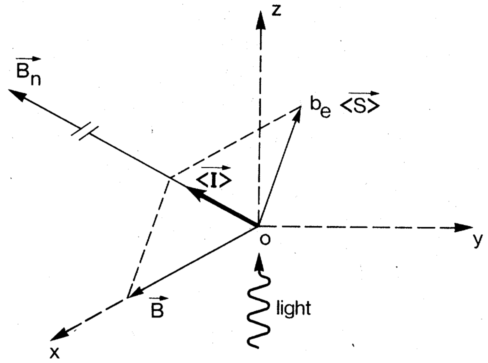

FIG. 1. Geometry of the system in a purely transverse external field: The field $\vec{B}$ is of the same order as the electronic field $b_e\langle\vec{S}\rangle$; the nuclei see a total oblique field which sets the direction of the nuclear spin $\langle\vec{I}\rangle$ and consequently of the large nuclear field $\vec{B}_n$. The electrons then experience a large depolarizing oblique magnetic field.

This shows that the usual optical pumping measurement of electronic lifetime and spin relaxation time from the Hanle curve is only possible when no nuclear effects are present or if they have been cancelled out.

IV. EXPERIMENTS

In our experiments, we measure the degree of circular polarization of the luminescence light $\mathcal{P}$. The experimental setup is shown on Fig. 2. The electronic spin is

$$\langle S_z\rangle=-\mathcal{P}\;. \tag{4.1}$$

The exciting beam from a krypton laser ($\lambda=7525$ Å; power 20 mW) is focused on the sample at 77 °K, the surface of the laser spot being 0.1 mm^2. The luminescence light is analyzed by a Spex 1700 monochromator, and the degree of circular polarization of the luminescence is detected with a rotating quarter-wave plate and synchronous detection. Care must be taken when performing the experiments, firstly by compensating all the parasitic fields, especially the earth field, secondly by

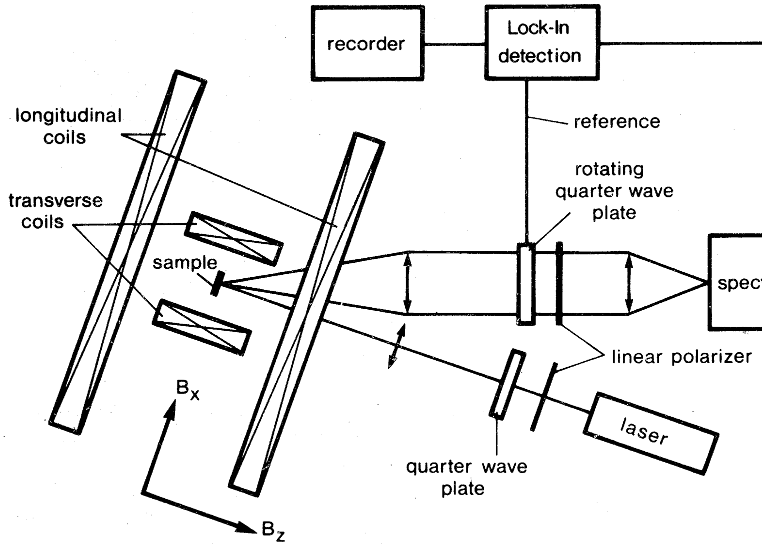

FIG. 2. Experimental setup: The parasitic fields have to be compensated and proper alignment of the direction of optical pumping (direction of the exciting laser) with the direction of the field configuration is of primary importance, as explained in the theory.

precisely aligning the light and the magnetic fields.

Alloys such as $Ga_xAl_{1-x}As$ exhibit strong nuclear effects in high fields[5] but these compounds are known to present important quadrupolar couplings which may modify the properties of the nuclear-spin system in low external fields. For this reason, we have chosen to work in a stoichiometric compound, gallium arsenide. Strong nuclear fields appear in pure GaAs at helium temperature,[7] but we have chosen highly doped and compensated samples for the following reasons: Intense luminescence, high degree of polarization, and a short nuclear relaxation time at 77 °K which makes possible the study of the electron-nuclear system in steady-state regime.[8,38] The samples used in these experiments are epitaxial p-type GaAs samples doped with Zn impurities ($N_A \simeq 2 \times 10^{18}$ cm^{-3}) and with various compensations by Te impurities. The degree of compensation varies from 0% to 50%. In these crystals, there are charge fluctuations due to the random distribution of ionized impurities and the carriers are localized in the resulting potential wells. This determines the transport and the optical properties (band tailing, kinetics of the recombination, luminescence, ...).[38–40] In spite of the complexity of the system, it can be shown that the electronic localization range is of the order of the donor Bohr radius[41] so that the numerical values of the electronic and nuclear fields given in Eqs. (2.13) and (2.19) are not too bad approximations. The luminescence spectrum of the samples is reduced to a single broad line around 8700 Å which is described in detail in Ref. 38.

We now present our measurements of the electronic polarization as a function of the applied magnetic field. We can either modulate the polarization of the exciting light or of the luminescence. At first sight, one could think that if the polarization of the exciting light is modulated faster than the total nuclear spin-lattice relaxation time, including the leak, the nuclear magnetization cannot build up. In fact this is not true in all cases, as shown in Ref. 12, where are discussed the nuclear effects obtained with simultaneous modulation of $\langle \vec{S}_0 \rangle$ and $\vec{B}$ faster than the nuclear spin-lattice relaxation time. Nevertheless in our case most of the nuclear effects diminish rapidly with increasing modulation frequency and totally disappear at 50 kHz.[42] The nuclear effects presented here are obtained for a stationary polarization of the exciting light. There are two simple experimental configurations depending on which component (transverse or longitudinal) of the external field is swept.

Figure 3 shows the results obtained by sweeping the longitudinal field through zero in a given transverse field: The general shape of the observed curves is in qualitative agreement with the theoretical prediction. The centers of these W-like curves do not correspond to zero-longitudinal field; there is a shift which is independent of the value of the static transverse field and which reverses its sign with the polarization of the exciting light. This shift is a direct measurement of the electronic field $\Gamma b_e \langle S_0 \rangle$

$$\Gamma b_e \langle S_0 \rangle = 0.9 \pm 0.1 \text{ G} \tag{4.2}$$

for σ^+ excitation. The quantity $\langle S_0 \rangle$ which has been defined as the electronic polarization in zero total field acting on the electrons is measured to be

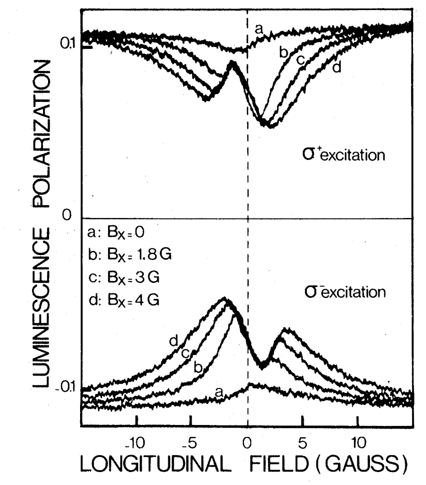

FIG. 3. Variation of the luminescence polarization for σ^+ and σ^- exciting light with the longitudinal field B_z for several values of the transverse magnetic field B_x: The shift of the center of the lines is equal to minus the electronic field. We measure $\Gamma b_e \langle S_0\rangle = 0.9 \pm 0.1$ G. The small variation of the electronic polarization when $B_x = 0$ (curve a) is due to the field dependence of the electronic relaxation time.

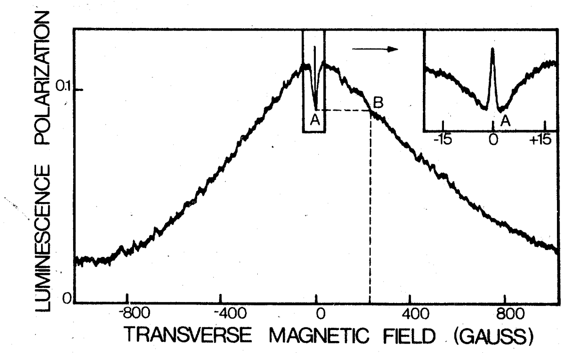

FIG. 4. Variation of the degree of circular polarization of the luminescence in a purely transverse magnetic field: When the transverse field is large the field seen by the nuclei is quasiperpendicular to the light excitation, the nuclei cannot get polarized and a usual Hanle depolarization curve is observed. In a field of the same order as the electronic field (insert) the total field seen by the nuclei is oblique and the electrons are depolarized by the transverse component of the nuclear field which can be as strong as 250 G (points A and B). In a zero transverse field, the nuclear field is longitudinal and has no depolarizing effect on the electrons.

$$\langle S_0\rangle = -0.10 \pm 0.01 . \tag{4.3}$$

In a purely longitudinal field, $\langle S_z\rangle$ should be constant and equal to $\langle S_0\rangle$. The small increase observed on Fig. 3(a) is attributed to the increase of the electronic relaxation time with the magnetic field seen by the electrons,[5] which in the present case is the nuclear field. An increase of the same order is observed in an external longitudinal field of 1 kG when the nuclear field is cancelled out by modulating the excitation light.

The transverse Hanle effect is presented in Fig. 4. For high values of the transverse magnetic field ($B_x > 20$ G) the total field seen by the nuclei is quasitransverse and the nuclei cannot get polarized: This large transverse field region reproduces a classical Hanle depolarization in the external field; the linewidth ΔB is found to be

$$\Delta B = 500 \pm 50 \text{ G} \tag{4.4}$$

This is no longer true in transverse fields of the order of the electronic field (see insert of Fig. 4). In this region, the nuclei see an oblique magnetic field and are polarized. The presence of the low-field depolarization of the luminescence light (W-like singularity) shows that in an external field of 2 G (point A of Fig. 4) one can obtain a transverse component of the nuclear field as high as 250 G (point B). These effects are due to the presence of the longitudinal component of the electronic field. They should disappear if one compensates the electronic field by a longitudinal external field of magnitude $B_z = -\Gamma b_e \langle S_0\rangle$. This is shown on Fig. 5. In fact the compensation is not perfect; this can be due to the previously mentioned field dependence of the electronic relaxation time, to spatial inhomogeneities of the electronic field or to inhomogeneities in the nuclear-spin system due to quadrupolar interactions.

We have observed that the nuclear field effects increase with the intensity of light excitation. This is probably due to the shortening of the nuclear relaxation time which increases the leakage factor f. The nuclear effects also increase with the compensation of the samples for the same reason and also because of increased localization. Quantitative comparison between theory and experiment can be made first from the transverse Hanle effect data given in Fig. 4. The quantity

$$\Lambda = \frac{B_x}{\Delta B}\,\frac{\langle S_z\rangle}{\langle S_0\rangle}\left(\frac{\langle S_0\rangle}{\langle S_z\rangle} - 1\right)^{1/2} \tag{4.5}$$

which can be obtained from the experiment is from Eqs. (3.41) and (3.42) equal to

$$\Lambda = \frac{B_x^2 + \xi B_L^2}{f b_n \Gamma b_e \langle S_0\rangle^2} \tag{4.6}$$

A plot of the measured value of Λ as a function of B_x^2 is shown in Fig. 6. The linear dependence on B_x^2 is observed and yields values of $f b_n \Gamma b_e \langle S_0\rangle^2$ and ξB_L^2. From the measured electronic field $\Gamma b_e \langle S_0\rangle$ we obtain the value of the nuclear field.

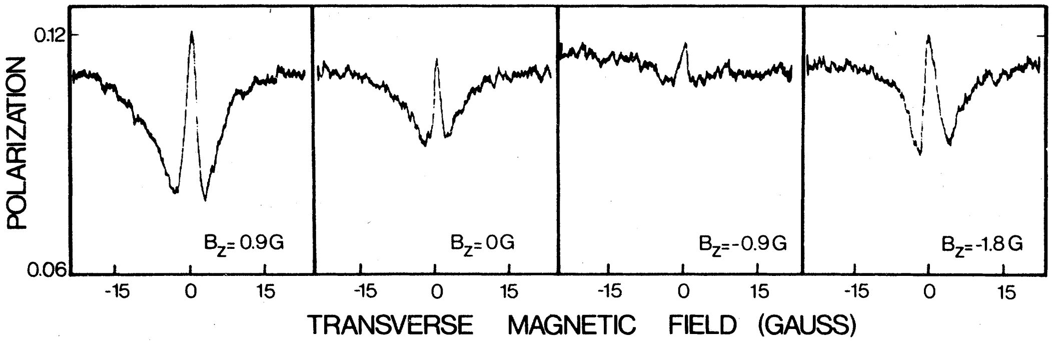

FIG. 5. Modification of the low-field singularities of the Hanle curve (Fig. 4) by addition of a small longitudinal field B_z: The singularity is strongly diminished when B_z is opposite to the electronic field so that the nuclei see a purely transverse field. It was not possible to achieve a perfect compensation. The best compensation of the electronic field is obtained for $B_z = -0.9$ G for σ^+ excitation in agreement with the longitudinal field results of Fig. 3.

Using these values it is possible to calculate from Eqs. (3.41) and (3.42) the electronic polarization for any given external field. This is shown in Fig. 7 which presents the comparison between theory and experiment in the longitudinal sweep case. Curves (a) and (b) are the experimental measurements of $\langle S_z\rangle$ as a function of B_z for two values of B_x: $B_x = 0$ and $B_x = 4$ G. Curve (c) is the theoretical variation of $\langle S_z\rangle$ given by Eqs. (3.41) and (3.42) for $B_x = 4$ G. The overall experimental behavior is reproduced. Better agreement can be obtained if we take into account the previously mentioned increase $\Delta\langle S_0\rangle = \langle S_z\rangle - \langle S_0\rangle$ of $\langle S_z\rangle$ with the external field B_z when $B_x = 0$. The corrected theoretical curve (d) is obtained by adding to curve (c) the small quantity $(\Delta\langle S_0\rangle)B_z^2/B^2$. (This can be justified in a first approximation by including in the Bloch-like equation (3.11) the field variation of T_1^e keeping T_2^e constant.) There remains a discrepancy between theory and experiment in the vicinity of $B_z = -\Gamma b_e\langle S_0\rangle$. This was already observed in our transverse field study (Fig. 5) and was attributed to the inhomogeneities of the system.

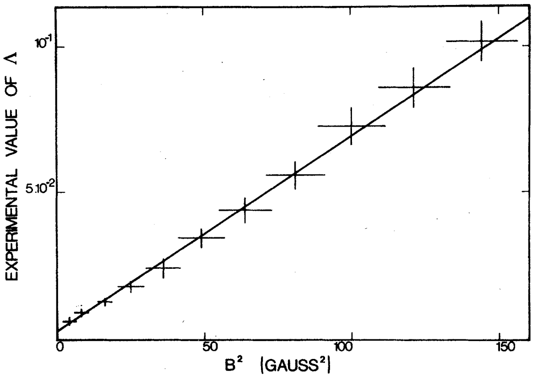

FIG. 6. Variation of the parameter Λ defined by Eq. (4.5) obtained from the Hanle effect data, as a function of the square of the transverse magnetic field B_x. As shown by Eq. (4.5) the dependence on B_x^2 should be linear. This is verified here. The slope gives $f b_n \Gamma b_e \langle S_0\rangle^2 = 1500 \pm 200$ G^2 the extrapolation to zero field yields $\xi B_L^2 = 6.2 \pm 1.0$ G^2.

V. DISCUSSION AND CONCLUSION

All our experimental results are correctly interpreted by a theoretical model which involves the mean electronic spin $\langle S_0\rangle$, the electronic field Γb_e, the nuclear field $f b_n$, and the quantity ξB_L^2.

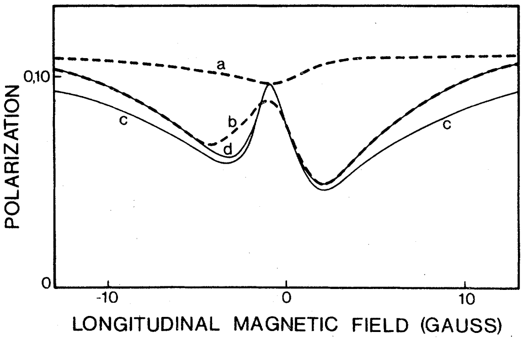

FIG. 7. Comparison between experiment and theory: Curves a and b are the experimental variation of the electronic polarization when the longitudinal field is swept through zero for two values of the transverse field: $B_x = 0$ and $B_x = 4$ G [Fig. 3(a) and 3(d)]. Curve c is the theoretical curve obtained from Eqs. (3.41) and (3.42) with $\langle S_0\rangle = -0.1$, $\Delta B = 500$ G, $f b_n = -17$ kG, and $\Gamma b_e = -9$ G. Curve d is the theoretical curve obtained from curve c after a first-order correction which takes into account the field dependence of the electronic relaxation time.

Their measured values are

$$\begin{aligned} f b_n &= -17 \pm 2 \text{ kG}, \\ \Gamma b_e &= -9 \pm 1 \text{ G}, \\ \xi B_L^2 &= 6.2 \pm 1.0 \text{ G}^2. \end{aligned} \tag{5.1}$$

These quantities are complicated averages depending on (i) the nuclear-spin system which has already been extensively studied, (ii) the electronic states which cannot be accurately described in our highly doped and highly compensated samples, and (iii) the mechanisms of nuclear relaxation which are not known in detail.

We have made theoretical estimations of these three quantities in a simplified case: The nuclear polarization is taken to be homogeneous near a center of localization for each of the three spin species and all quadrupolar effects are neglected. The inclusion of the quadrupolar interaction implies the description of the local electric field gradients which depend on the impurities and cannot simply be approximated in highly doped samples. They create inhomogeneities in the nuclear-spin system, and the fluctuating recombination of electrons on a given center constitutes for the nearby nuclei a leak mechanism correlated with the polarizing mechanism. This may not be negligible and further studies are necessary to include the quadrupolar effects. The overall agreement of our experiments in low field with our model shows nevertheless that they do not drastically alter our results. The effect of the quadrupolar interactions is probably to modify the actual values of the parameters and we shall not try to compare them precisely with the theoretical estimations. In the following we discuss the experimental determinations of $f b_n$, Γb_e, and ξB_L^2.

The experimental nuclear field $f b_n = -17 \pm 2$ kG is one order of magnitude smaller than the theoretical nuclear field $b_n = -170$ kG. This theoretical value is calculated in the Appendix supposing that the nuclear polarization of each spin species is homogeneous in space. It depends on the values d_α of the electronic wave functions on the nuclei. These quantities have been estimated in Appendix A and we are confident in their numerical values. Thus we find the value of the leakage factor $f \sim 0.1$. This factor is the ratio of the nuclear relaxation rate due to the photoelectrons $(T_{1e})^{-1}$ to the total nuclear-relaxation rate $(T_{1e})^{-1} + (T_{1f})^{-1}$. This total rate can be estimated in our experimental conditions to be of the order of a fraction of a second: No transient effects on a scale longer than 1 sec have been observed, and on the other hand, the nuclear effects are diminished when the excitation light is modulated at 70 Hz which shows that the total nuclear relaxation time is longer than 10^{-2} sec. Thus the relaxation time due to the photoelectrons is of the order of a fraction of a second. It appears to be in agreement with the theoretical estimate given by Eq. (2.15) provided that the occupation factor Γ_t is not too small. The relaxation time due to the other mechanisms of nuclear relaxation is shorter than the relaxation times due to phonon-quadrupolar couplings which have been measured in GaAs to be of the order of tens of seconds at 77 °K.[23] The leak mechanism in our strongly doped p-type samples may be due to the hyperfine interaction of the nuclei with the holes. Let us note, however, that from the value of the leakage factor we cannot draw definite conclusions on the nuclear-relaxation mechanisms because the static quadrupolar effect could be partially responsible for the smaller values of the nuclear magnetizations.

A comparison of the value of the electronic field $\Gamma b_e = -9$ G with the theoretical estimate given by Eq. (3.36): $b_e = -170$ G shows that the quantity $\Gamma = \Gamma_s \Gamma_t$ is of the order of 0.05 in experimental conditions where there exist approximately 10^{15} photoelectrons per cm^3. If all the electrons were trapped indifferently on the impurities (of concentration 10^{18} cm^{-3}) one would expect the proportion of occupied centers Γ_t to be of the order of 10^{-3} and $\Gamma_s \Gamma_t$ should be even smaller because the spatial average Γ_s of the electronic wave function is smaller than one. The very high experimental value of the product $\Gamma_s \Gamma_t$ indicates that the photoelectrons are preferentially trapped on a small fraction of the localization centers, probably those of lower energy, and the observed nuclei are situated near these centers.

We discuss now the experimental value of the spin-spin parameter: $\xi B_L^2 = 6.24 \pm 1.0$ G^2. As said before, the local field B_L can be calculated in GaAs from NMR and NAR linewidths. The calculation is given in Appendix B: $B_L^2 = 2.1 \pm 0.1$ G^2. We also show in Appendix B that the factor ξ is essentially determined by the ratio of dipolar plus pseudodipolar fields to the total local field which includes the exchange fields. For this reason ξB_L^2 can be considered to be known in GaAs almost as well as B_L^2. The theoretical prediction ξB_L^2 is 4.6 ± 0.5 G^2; there is a good agreement between experiment and theory.

The quantitative agreement between theory and experiment and the fact that the measured physical parameters are comparable to values derived from first principles lead to the following conclusions. We understand the basic physical effects involved in low-field nuclear optical pumping phenomena, at least in a steady-state regime with not too large an electronic field. This is exemplified by the fact that the optical-pumping determination of the quantity ξB_L^2 coincides with the determination of this quantity from nuclear magnetic and nuclear acoustic resonance in bulk gallium arsenide. The basis established in this work which deals with the nuclear-spin temperature concept in the laboratory frame can be extended to optical detection of NMR in the rotating frame.

The usual optical-pumping determination of the electronic relaxation time and lifetime from the values of the Hanle linewidth has to be done with special care whenever nuclear effects are present, for example, by modulation of the light excitation, transient measurements or saturation of the nuclear magnetization, etc.

The measurement of luminescence polarization as a function of a longitudinal field in a static transverse field gives a direct determination of the electronic field Γb_e. A more detailed analysis yields the values of the nuclear field $f b_n$ and of the nuclear spin-spin interaction ξB_L^2. Let us note that in doped and compensated GaAs there is only one line in the recombination spectra but the same measurements of the electronic and nuclear fields can be done in principle on each line of a complex luminescence spectrum. This may give the values of the electronic and nuclear fields in each of the excited states of the crystal (free or bound excitons, donors and acceptors, etc.) yielding detailed informations about wave functions and dynamics of recombination.

APPENDIX A: EVALUATION OF THE ELECTRONIC WAVE FUNCTION AT THE POSITION OF THE NUCLEI

Hyperfine interactions have not been measured in gallium arsenide but it is possible to scale their values from experimental data in indium antimonide.[17] Our scaling is based on the fact that gallium arsenide and indium antimonide have very close ionicities (respectively 0.310 and 0.321 in the Philips scale).[43] This means that the sharing of the electrons between In and Sb atoms in InSb is the same as between Ga and As atoms in GaAs. Although this is related to the valence (bonding) states, the same property should hold in first approximation for the conduction (antibonding) states: We assume that this is true for the $\vec{k}=0$ state. Then the ratio of the hyperfine interactions from GaAs to InSb is the same as the ratio of the *atomic* hyperfine interactions, and

$$\frac{d_{(\mathrm{Ga}\ in\ \mathrm{GaAs})}}{d_{(\mathrm{In}\ in\ \mathrm{InSb})}}=\frac{|\psi(0)|^2_{(\mathrm{Ga}\ atom)}}{|\psi(0)|^2_{(\mathrm{In}\ atom)}}, \tag{A1}$$

$$\frac{d_{(\mathrm{As}\ in\ \mathrm{GaAs})}}{d_{(\mathrm{Sb}\ in\ \mathrm{InSb})}}=\frac{|\psi(0)|^2_{(\mathrm{As}\ atom)}}{|\psi(0)|^2_{(\mathrm{Sb}\ atom)}}. \tag{A2}$$

The atomic hyperfine-structure values which appear on the right-hand side of the above equation are known.[44] The quantities $d_{(\mathrm{In}\ in\ \mathrm{InSb})}$ and $d_{(\mathrm{Sb}\ in\ \mathrm{InSb})}$ have been measured by Guéron.[17] One obtains then,

$$d_{(\mathrm{Ga}\ in\ \mathrm{GaAs})}\simeq 5.8\times 10^{25}\ \mathrm{cm}^{-3}, \tag{A3}$$

$$d_{(\mathrm{As}\ in\ \mathrm{GaAs})}\simeq 9.8\times 10^{25}\ \mathrm{cm}^{-3}. \tag{A4}$$

APPENDIX B: CALCULATION OF THE NUCLEAR FIELD

We start from Eq. (3.30) and we express that in steady-state regime the total energy of the system is time independent. This leads to

$$0=\sum_{\mu}\mathrm{Tr}(\sigma-\sigma_\mu)\int_0^\infty\langle\{\mathcal{H}'_\mu(t),[\mathcal{H}'_\mu(t-\tau),\mathcal{H}_0]\}\rangle\,d\tau. \tag{B1}$$

We suppose that the relaxing interactions are scalar, i.e., of the form

$$\mathcal{H}'_\mu(t)=-\sum_i\gamma_i\hbar\vec{\mathrm{B}}^i_\mu(t)\cdot\vec{\mathrm{I}}_i, \tag{B2}$$

where the nuclear spin $\vec{\mathrm{I}}_i$ of gyromagnetic factor γ_i experiences a fluctuating field $\vec{\mathrm{B}}^i_\mu(t)$. The field $\vec{\mathrm{B}}^i_e(t)$ is the fluctuating part of the field $\vec{\mathrm{B}}^i_e$ defined by Eq. (2.5). (We recall that the static part $\langle\vec{\mathrm{B}}^i_e\rangle$ of $\vec{\mathrm{B}}^i_e$ is incorporated into $\mathcal{H}_0$.) The static Hamiltonian $\mathcal{H}_0$ of the nuclear spin system is given by Eqs. (3.15)–(3.20). The quantities $\mathcal{B}_{ij}$ and a_{ij} which appear in Eqs. (3.18) and (3.19) have been measured in GaAs by NMR and NAR experiments.[30,37] The number $\mathcal{B}_{ij}$ is nonzero only when i and j are nearest neighbors (Ga and As) and is the same for ^{69}Ga–^{75}As and ^{71}Ga–^{75}As

$$(1+\mathcal{B}_{^{69}\mathrm{Ga}^{75}\mathrm{As}})^2=(1+\mathcal{B}_{^{71}\mathrm{Ga}^{75}\mathrm{As}})^2=1.64\pm 0.13. \tag{B3}$$

The quantity a_{ij} is independent of the isotopic species and in the particular case of GaAs, independent of the chemical species ($a_{ij}=a$)

$$a^2=(2.6\pm 0.3)\times 10^{-16}\ \mathrm{cm}^2 \tag{B4}$$

The calculation of the double commutators of Eq. (B1), knowing the expression of $\mathcal{H}_0$, is somewhat tedious. They involve the correlation functions $\langle B^i_{\mu m}(t)B^j_{\mu n}(t-\tau)\rangle$ where m and n stand for x, y or z. We suppose (a) that the interactions $\mathcal{H}'_f(t)$ and $\mathcal{H}'_e(t)$ are uncorrelated (b) that the fields $\vec{\mathrm{B}}^i_\mu(t)$ and $\vec{\mathrm{B}}^j_\mu(t)$ are totally correlated for neighboring nuclei. This is reasonable for the polarizing field $\vec{\mathrm{B}}^i_e(t)$ because of the small value of the interatomic distance as compared with the extension of the electronic state. The same hypothesis is made for the depolarizing fields $\vec{\mathrm{B}}^i_f(t)$. The correlation functions are taken as

$$\langle B^i_{\mu m}(t)B^j_{\mu n}(t-\tau)\rangle=\tfrac{1}{3}\delta_{mn}b^{ij}_\mu\exp(-\tau/\tau_\mu), \tag{B5}$$

where

$$b_\mu^{ij} = \langle \vec{B}_\mu^i(t=0) \cdot \vec{B}_\mu^j(t=0) \rangle , \qquad \text{(B6)}$$

and where the τ_μ's are the correlation times of the interactions $\mathcal{H}'_\mu(t)$ (of the order of nanoseconds).

Then, using the assumptions made above, Eqs. (B1), (B2), and (B5) and the expression (3.14) of the density matrix we obtain the steady-state value of the nuclear spin temperature θ

$$\frac{1}{k_B\theta}\sum_\mu \frac{2}{3}\tau_\mu \left(\sum_i b_\mu^{ii}\gamma_i^2 \frac{\mathrm{Tr} Z_i^2}{\mathrm{Tr}\underline{I}} + \sum_{i>j} \frac{\mathrm{Tr}(1+\mathcal{B}_{ij})^2 \mathcal{H}_{Dij}^2}{\mathrm{Tr}\underline{I}} (b_\mu^{ii}\gamma_i^2 + b_\mu^{jj}\gamma_j^2 + b_\mu^{ij}\gamma_i\gamma_j) + \sum_{i>j}\frac{\mathrm{Tr}\mathcal{H}_{Eij}^2}{\mathrm{Tr}\underline{I}}(b_\mu^{ii}\gamma_i^2 + b_\mu^{jj}\gamma_j^2 - 2b_\mu^{ij}\gamma_i\gamma_j)\right)$$

$$= \frac{2}{3}\tau_e \sum_i (b_e^{ii}\gamma_i^2)(\gamma_i\hbar)\frac{3\,\mathrm{Tr}(\vec{I}_i\cdot\langle\vec{S}\rangle)[(\vec{B}+\langle\vec{B}_e^i\rangle)\cdot\vec{I}_i]}{(\mathrm{Tr}\underline{I})\,S(S+1)} . \qquad \text{(B7)}$$

The Zeeman energies Z_i are given by Eq. (3.15). In the case of an inhomogeneous system with several species of nuclear spins experiencing a distribution of relaxing fields, the sums of Eq. (B7) can be averaged. But, as said in the text, we restrict ourselves to the case where all the spins α experience the same relaxing fields. The quantities b_μ^{ij} are then equal to $b_\mu^{\alpha\beta}$ for all the spins i of species α and all the spins j of species β.

To simplify the expression of θ we introduce the partial local fields $B_{D\alpha\beta}$ and $B_{E\alpha\beta}$ which are respectively dipolar (including pseudodipolar) and exchange local fields between spins of species α and β:

$$B_{D\alpha\beta}^2 = \mathrm{Tr}\sum_{i\in\alpha,\, j\in\beta,\, i>j} (1+\mathcal{B}_{ij})^2\mathcal{H}_{Dij}^2 \times\left(\tfrac{1}{3}\,\mathrm{Tr}\sum_\alpha n_\alpha\gamma_\alpha^2\hbar^2 I_\alpha^2\right)^{-1} , \qquad \text{(B8)}$$

$$B_{E\alpha\beta}^2 = \mathrm{Tr}\sum_{i\in\alpha,\, j\in\beta,\, i>j} \mathcal{H}_{Eij}^2\left(\tfrac{1}{3}\,\mathrm{Tr}\sum_\alpha n_\alpha\gamma_\alpha^2\hbar^2 I_\alpha^2\right)^{-1} . \qquad \text{(B9)}$$

The local field defined in Eq. (3.27) is equal to

$$B_L^2 = \tfrac{1}{2}\sum_{\alpha,\beta}(B_{D\alpha\beta}^2 + B_{E\alpha\beta}^2) . \qquad \text{(B10)}$$

It is also convenient to introduce the different relaxation times of the various interactions: The spin relaxation time $1/T_1^\alpha$ of the Zeeman energy

$$1/T_1^\alpha = 1/T_{1e}^\alpha + 1/T_{1f}^\alpha , \qquad \text{(B11)}$$

with

$$1/T_{1e}^\alpha = \tfrac{2}{3}\, b_e^{\alpha\alpha}\gamma_\alpha^2\tau_e , \qquad \text{(B12)}$$

$$1/T_{1f}^\alpha = \tfrac{2}{3}\, b_f^{\alpha\alpha}\gamma_\alpha^2\tau_f . \qquad \text{(B13)}$$

The spin-relaxation time $1/T_1^{D\alpha\beta}$ of dipolar plus pseudodipolar energies:

$$\frac{1}{T_1^{D\alpha\beta}} = \left[\frac{1}{T_{1e}^\alpha} + \frac{1}{T_{1e}^\beta} + \left(\frac{1}{T_{1e}^\alpha}\frac{1}{T_{1e}^\beta}\right)^{1/2}\right] + \left[\frac{1}{T_{1f}^\alpha} + \frac{1}{T_{1f}^\beta} + \left(\frac{1}{T_{1f}^\alpha}\frac{1}{T_{1f}^\beta}\right)^{1/2}\right] . \qquad \text{(B14)}$$

The spin-relaxation time $1/T_1^{E\alpha\beta}$ of the exchange energy:

$$\frac{1}{T_1^{E\alpha\beta}} = \left[\frac{1}{T_{1e}^\alpha} + \frac{1}{T_{1e}^\beta} - 2\left(\frac{1}{T_{1e}^\alpha}\frac{1}{T_{1e}^\beta}\right)^{1/2}\right] + \left[\frac{1}{T_{1f}^\alpha} + \frac{1}{T_{1f}^\beta} - 2\left(\frac{1}{T_{1f}^\alpha}\frac{1}{T_{1f}^\beta}\right)^{1/2}\right] . \qquad \text{(B15)}$$

The rate of relaxation of the exchange energy between like spins $1/T_1^{E\alpha\alpha}$ is zero. This means that the exchange energy between like spins is not relaxed by scalar interactions, a well known result.

With these definitions, the nuclear-spin temperature is given by

$$\frac{1}{k_B\theta} = \frac{3}{S(S+1)}\;\frac{\langle\vec{S}\rangle\cdot\sum_\alpha \vec{B}_T^\alpha \frac{c_\alpha}{T_{1e}^\alpha}\frac{1}{\gamma_\alpha\hbar}}{\left[\sum_\alpha \frac{c_\alpha}{T_1^\alpha}(B_T^\alpha)^2 + \frac{1}{2}\sum_{\alpha,\beta}\left(\frac{1}{T_1^{D\alpha\beta}}B_{D\alpha\beta}^2 + \frac{1}{T_1^{E\alpha\beta}}B_{E\alpha\beta}^2\right)\right]} . \qquad \text{(B16)}$$

The quantity c_α is defined by Eq. (3.26). We have calculated the quantities $B_{D\alpha\beta}^2$ and $B_{E\alpha\beta}^2$ for GaAs (zinc-blende structure, lattice parameter equal to 5.65 Å). Their values are given in Table I. The resulting value of the local field is

$$B_L^2 = 2.1 \pm 0.1\ \mathrm{G}^2 . \qquad \text{(B17)}$$

As seen on Table I the contributions of the exchange interaction is about one fourth of this value.

The nuclear field $\vec{B}_\alpha$ is then calculated from Eq. (2.17) and Eq. (3.24). If in the denominator of Eq. (B16) we neglect the electronic field as compared to the local field, we obtain[45]:

$$\vec{B}_\alpha = \left(\sum_{\alpha'}\frac{T_1}{T_{1e}^{\alpha'}}\frac{c_{\alpha'}}{\gamma_{\alpha'}}(\vec{B}_T^{\alpha'}\cdot\langle\vec{S}\rangle)\right)\frac{I_\alpha(I_\alpha+1)}{S(S+1)}\frac{\gamma_\alpha b_\alpha}{B^2+\xi B_L^2}\vec{B}_T^\alpha , \qquad \text{(B18)}$$

where

TABLE I. Magnitudes of the various contributions to the local field (in units of 10^{-2} G^2). The table gives the contribution of dipolar plus pseudodipolar $B^2_{D\alpha\beta}$ and exchange $B^2_{E\alpha\beta}$ interactions between the nuclear-spin species α and β, calculated from Eqs. (B8) and (B9). Note that the dipolar local fields between like spins do not depend on pseudodipolar interactions and can be calculated very precisely.

$\alpha\,\beta$	^{69}Ga^{69}Ga	^{71}Ga^{71}Ga	^{75}As^{75}As	^{69}Ga^{71}Ga	^{71}Ga^{75}As	^{75}As^{69}Ga
$B^2_{D\alpha\beta}$	11	13	10	23	51 ± 4	47 ± 4
$B^2_{E\alpha\beta}$	0.66 ± 0.1	1.0 ± 0.1	0.66 ± 0.1	1.7 ± 0.3	26 ± 3	24 ± 3

$$1/T_1 = 1/T_{1e} + 1/T_{1f}, \tag{B19}$$

$$\frac{1}{T_{1e}} = \sum_\alpha \frac{c_\alpha}{T^\alpha_{1e}}, \tag{B20}$$

$$\frac{1}{T_{1f}} = \sum_\alpha \frac{c_\alpha}{T^\alpha_{1f}}, \tag{B21}$$

and

$$\xi = \frac{T_1}{2B_L^2} \sum_{\alpha,\beta} \left(\frac{B^2_{D\alpha\beta}}{T_1^{D\alpha\beta}} + \frac{B^2_{E\alpha\beta}}{T_1^{E\alpha\beta}} \right). \tag{B22}$$

As pointed out in the discussion of Eq. (2.12) we need only to consider the part of the fields $\vec{B}_\alpha$'s along $\vec{B}$ to obtain the effective nuclear field $\vec{B}'_N$. It is convenient to define an "equivalent leakage factor" f, weighted gyromagnetic ratios $\tilde{\gamma}_\alpha$ and $\tilde{\gamma}$, and also an "equivalent nuclear field" b_n and an "equivalent electronic field" b_e

$$f = T_1/T_{1e}, \tag{B23}$$

$$\frac{1}{\tilde{\gamma}_\alpha} = \frac{c_\alpha}{\gamma_\alpha} \frac{T_{1e}}{T^\alpha_{1e}}, \tag{B24}$$

$$\frac{1}{\tilde{\gamma}} = \sum_\alpha \frac{1}{\tilde{\gamma}_\alpha}, \tag{B25}$$

$$b_n = \sum_\alpha \frac{I_\alpha(I_\alpha+1)}{S(S+1)} \frac{\gamma_\alpha}{\tilde{\gamma}} b_\alpha, \tag{B26}$$

$$b_e = \sum_\alpha \frac{\tilde{\gamma}}{\tilde{\gamma}_\alpha} b^\alpha_e(0). \tag{B27}$$

Using these quantities, Eqs. (2.2) and (2.20) and Eq. (B18) one obtains finally

$$\vec{B}'_N = f b_n \frac{\langle\vec{S}\rangle \cdot (\vec{B} + \Gamma b_e \langle\vec{S}\rangle)}{B^2 + \xi B_L^2} \vec{B} \tag{B28}$$

The quantities $\tilde{\gamma}_\alpha$ can be expressed only in terms of γ_α, x_α, and d_α as can be seen from Eqs. (B6), (B12), and (B20) and Eqs. (2.11) and (2.12). From the numerical values (2.13) and (2.19) of $b^\alpha_e(0)$ and b_α one obtains

$$b_n = -170 \text{ kG}, \tag{B29}$$

$$b_e = -170 \text{ G}. \tag{B30}$$

In principle, to calculate ξ one needs to know the nuclear relaxation mechanisms. In fact, ξ depends mainly on the relative importance of the dipolar and exchange local fields whereas it is only weakly sensitive to the details of the relaxation mechanisms. For this reason, we do not make a significant error by assuming that

$$T^\alpha_1/T^\alpha_{1e} = T^\beta_1/T^\beta_{1e} = f. \tag{B31}$$

This supposes that for each nuclear species the depolarizing interaction is proportional to the hyperfine interaction. In this case, the expression of ξ is greatly simplified and involves only known nuclear parameters:

$$\xi = \left(\sum_{\alpha,\beta} B^2_{D\alpha\beta}(\gamma^2_\alpha d^2_\alpha + \gamma^2_\beta d^2_\beta + \gamma_\alpha \gamma_\beta d_\alpha d_\beta) + \sum_{\alpha,\beta} B^2_{E\alpha\beta}(\gamma^2_\alpha d^2_\alpha + \gamma^2_\beta d^2_\beta - 2\gamma_\alpha \gamma_\beta d_\alpha d_\beta) \right) \bigg/ \left(2B_L^2 \sum_\alpha c_\alpha \gamma^2_\alpha d^2_\alpha \right) \tag{B32}$$

If only dipolar or pseudodipolar interactions were present in the crystal the value of ξ would be equal to 3 for a system with only one spin species and of the order of 3 when there exist several spin species, a well known result.[32] In our case where the exchange interactions are not negligible as compared to the other interactions (see Table I) the value of ξ is reduced. In practice, only the first sum of Eq. (B32) is important. The calculation yields

$$\xi = 2.2 \pm 0.2, \tag{B33}$$

and the value of the spin-spin parameter is

$$\xi B_L^2 = 4.6 \pm 0.5 \text{ G}^2. \tag{B34}$$

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