Magnetic-field dependence of the optical Overhauser effect in GaAs

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When nuclear spin order is induced by optical excitation near the band gap of a semiconductor such as GaAs, the effect is referred to as optical pumping. This paper presents measurements of the optical pumping rate in semi-insulating GaAs over the magnetic field range of $0-24$ T at temperatures of 1.5 K and 4.2 K. The enhanced nuclear polarization was sampled by radio wave detected NMR. The data were recorded using Bitter-type magnets which permitted rapid ramping between the pumping and sampling fields in a time short compared to the nuclear spin lattice relaxation time in the dark. The field dependence has been fitted to a relaxation model which includes spin diffusion and dark relaxation terms. Fits were obtained by fixing the g factor to its literature value. The fitted parameters include the correlation time for electron spin-density fluctuations, the average hyperfine field, and the nuclear spin diffusion coefficient. [S0163-1829(97)02112-7]

I. INTRODUCTION

NMR Knight shift measurements in $Al_xGa_{1-x}As/GaAs$ quantum wells have recently provided dramatic confirmation for the existence of certain novel quantum states in the quantum Hall effect.[1,2] The measurements were facilitated by optical pumping, an effect whereby enhanced NMR sensitivity can be obtained by nuclear spin cross relaxation with optically oriented electrons. These important results provide the impetus to characterize the field dependence of the optical pumping effect. Here, the theory is extended to arbitrarily high field in the simplest possible system: bulk, semi-insulating GaAs. The theory is compared with the experimental field dependence over the range of $0-24$ T. In the experiments described here, the nuclear spin polarization was sampled directly by radio wave NMR detection in a tuned coil. The field dependence is fitted to a relaxation model which includes spin diffusion and dark relaxation terms. Simplex fits to the data provide the values for several key parameters that govern the electron-nuclear relaxation and steady state spin polarizations.

II. OPTICAL PUMPING AND THE OPTICAL OVERHAUSER EFFECT

At low temperature, nuclear spin lattice relaxation in a semiconductor can be dominated by fluctuations in the hyperfine contact interaction with localized electrons.[3] In bulk GaAs the source of these fluctuations has been identified as spin exchange between delocalized electrons and electrons trapped at shallow donor impurities.[4] Lampel originally demonstrated that hyperfine cross relaxation between nuclei and optically oriented conduction electrons trapped at point defects could dynamically polarize the ^{29}Si nuclei in bulk silicon.[5] Depending on the sample and experimental conditions, nuclear spin polarizations approaching unity may be obtained, leading to NMR signal enhancements of up to 10^4. This optical pumping effect occurs not only with circularly polarized light, but also with linear or unpolarized light in a manner analogous to the Overhauser effect.[6] Pumping with either unpolarized or plane polarized light is therefore referred to as the *optical Overhauser effect.*

In Lampel's experiment, the enhanced nuclear polarization was observed simply by radio wave (RF) detection with a tuned coil. Subsequently, Ekimov and Safarov[7] observed that the degree of polarization of the luminescence was altered by tilting the local nuclear fields by means of resonant NMR transitions. This optically detected NMR (ODNMR) technique has the principal advantage of extremely high sensitivity. In the context of ODNMR, the electron nuclear interactions in many types of bulk semiconductors have been studied in great detail. The high sensitivity of ODNMR has also enabled studies of single heterostructures.[8] Although this technique has higher sensitivity compared to radio wave detection, a basic limitation of ODNMR stems from the fact that it relies on changes in the luminescence depolarization due to the nuclear Hanle effect.[9] The local nuclear field, B_N, causes the quantization axis of the electron spin system to be tilted away from the external magnetic field B_0 by an angle $\tan^{-1}(B_N/B_0)$. Outside the Hanle regime, where $B_0 \gg B_N$, this angle approaches zero and the electron spin depolarization by the nuclear field is not effective. Thus, if the magnetic field dependent physics of interest occurs at relatively high field, ODNMR might not be applicable unless a B_0 cycling scheme is employed.

Here, theory and experiment are extended to high magnetic field. A model for the magnetic field dependence of optical pumping is constructed and solved in terms of time and the spatial displacement from the donor origin. Of particular interest is the efficiency of optical pumping at very high magnetic field. Variable field measurements were performed using a field cycling procedure on the Bitter magnets at NHMFL/Tallahassee. Ramp rates as high as 0.6 T/s are feasible on these resistive magnets. From the field dependence of the optically pumped NMR signal a number of relevant parameters can be extracted, namely the correlation

time τ_e of the electron spin and the coefficient of nuclear spin diffusion, D, and the average hyperfine field. The electron-nuclear cross relaxation due to the random variation of the hyperfine coupling fits the criteria for scalar relaxation of the first kind.[10] This formalism is used to explain the magnetic field variation of the optical pumping rate in GaAs. Simplex fits to the magnetic-field dependence data are presented.

III. THEORY

Selective optical excitation of electrons residing in the heavy-hole and/or light-hole valence-band states at energies near the band-gap creates conduction electrons that may become trapped near shallow donor sites. If the conduction electron spin relaxation time is of the electron in the conduction band during the excited state lifetime, then the average electron spin occupying the conduction band is determined by the interband dipole transition matrix elements. When expressed in terms of the retardation Γ between the ordinary and extraordinary electric field components, $\langle S_z\rangle = \sin(\Gamma)/4$. The average conduction electron spin can be diminished if reorientation of the electron occurs on a time scale short compared to the excited state lifetime. In terms of the transition rates r_+ and r_- to the $|+\rangle$ and $|-\rangle$ spin states, $\langle S_z\rangle$ is given by

$$\langle S_z\rangle = \frac{1}{2}\left(\frac{r_+ - r_-}{r_+ + r_-}\right)\frac{1}{1+\tau_e/T_{1S}} \tag{1}$$

$$= \frac{1}{4}\sin(\Gamma)\,\frac{1}{1+\tau_e/T_{1S}}, \tag{2}$$

where T_{1S} is the electron spin lattice relaxation time and τ_e is the excited state lifetime. Hence, the spin polarization of the donor-bound electrons can be determined by the ratio of the spin lattice relaxation time and the excited state lifetime, T_{1S}/τ_e. In GaAs, $T_{1S} \ll \tau_e$ at zero field.[9] The relaxation rate is greatly decreased by an applied field according to[3,9]

$$T_{1S}(H_0) = T_{1S}(0)\left[1+\left(\frac{B_0 \pm \eta B_N}{H_\gamma}\right)^2\right], \tag{3}$$

where $H_\gamma = A(g_e\mu_0)^{-1}\gamma$, γ is the frequency of the variation of the local fields acting on the electron, g_e is the effective g factor of the electron, and $T_{1S}(0)$ is the zero field spin lattice relaxation time of the electron. The factors A and η are constants. The field B_N is the total local nuclear field and is proportional to $\langle I_z\rangle$ summed over all isotopes. The nuclear field can either add to or subtract from the external field B_0 to shift the electron spin relaxation time to larger or smaller values. In GaAs, the condition $T_{1S} \gg \tau_e$ is approximately obtained at $B_0 \geq 1$ T at 4.2 K.[9] At this or lower temperatures, the electron spin polarization is therefore determined solely by the interband dipole transition matrix elements at higher magnetic fields (note that since the energy of the exciting light is close enough to the free exciton band gap, hot electron spin relaxation mechanisms can be neglected). Provided that $T_{1S} \gg \tau_e$, for light of positive (σ^+) helicity $\langle S_z\rangle = +1/4$, for negative ($\sigma^-$) helicity $\langle S_z\rangle = -1/4$, and for linear or unpolarized light $\langle S_z\rangle = 0$.

Dynamic polarization of nuclei by optically oriented electrons is known to occur in the vicinity of shallow donor sites where the electronic wave function can be described by the (normalized) ''effective-mass method'' wave function $\psi(r) = e^{-r/a_0}/\sqrt{\pi a_0{}^3}$, where $r=0$ describes the origin of the donor site and a_0 is the Bohr radius of the bound electron. In GaAs, $a_0 \approx 100$ Å.[3,9] The isotropic hyperfine interaction between the ith electron and a given nucleus at r in the donor complex is given by analogy with the interaction in the free atom,

$$\hat{\mathcal{H}}_{IS} = -a_{\rm Ni}\hat{\rm I}\cdot\hat{\rm S}_i, \tag{4}$$

where

$$a_{\rm Ni} = \frac{16\pi}{3}\mu_B\hbar\,\gamma_n v_0 |\psi(r)|^2 \tag{5}$$

is the hyperfine contact coupling constant for a nucleus situated at a displacement r with respect to the electron at the origin and v_0 is the unit cell volume. The anisotropic component of the hyperfine interaction should be small and will be neglected. The total Hamiltonian for an individual electron-nuclear spin pair is

$$\hat{\mathcal{H}} = \omega_S\hat{\rm S}_z + \omega_I\hat{\rm I}_z - a_{\rm Ni}(t)\hat{\rm I}\cdot\hat{\rm S}, \tag{6}$$

where ω_S and ω_I are the Larmor frequencies of the electron and nucleus, respectively.

Electron-nuclear cross relaxation is induced by random fluctuations in the spin density due to spin exchange with free electrons.[3] A simple physical model can be constructed if it is assumed that the coupling $a_{\rm Ni}(t)$ may take on either of two values: $a_{\rm Ni}$ during the time an electron is trapped at the donor and zero otherwise. The mean squared coupling can then be written as $\overline{[a_{\rm Ni}^2(t)]} = P_i a_N^2$, where P_i is the probability of finding the ith electron trapped on the donor in the vicinity of the nucleus. The correlation function of $a_{\rm Ni}(t)$ in this simple model is[10]

$$\overline{a_{\rm Ni}(t)\cdot a_{\rm Ni}(t+\tau)} = [\overline{a_{\rm Ni}(t)}]^2 e^{-\gamma_e\tau}. \tag{7}$$

The fluctuating field experienced by the nuclei may be characterized by a correlation frequency[3,4] $\gamma_e \approx 10^{11}$ s^{-1}. It is also evident that the magnitude of the fluctuations is maximal near the origin of the donor and the cross relaxation rate will therefore be maximal at $r=0$.

The equation for dipolar or scalar relaxation of the first kind can be derived from the density operator master equation. Following Abragam,[10]

$$\frac{d\langle I_z\rangle}{dt} = \frac{-1}{T_1^{II}}\left\{\langle I_z\rangle - I_0 + \zeta\frac{I(I+1)}{S(S+1)}(\langle S_z\rangle - S_0)\right\}. \tag{8}$$

For scalar relaxation of the first kind, $\zeta = -1$ (for dipolar hyperfine relaxation, $\zeta = +1/2$) and

$$\frac{1}{T_1^{II}} = \frac{S(S+1)}{3}J_{\rm exch}(\omega_I - \omega_S), \tag{9}$$

$$J_{\rm exch}(\omega) = \sum_i P_i \int_{-\infty}^{+\infty} e^{-i\omega\tau} d\tau\, \overline{a_{\rm Ni}(t)\cdot a_{\rm Ni}(t+\tau)}. \tag{10}$$

In Eq. (8) the expectation value of the electron spin at thermal equilibrium is given by

$$S_0 = -\frac{1}{2}\tanh\left(\frac{\hbar g_e \mu_0 B_0}{2kT}\right). \tag{11}$$

The probability that an electron is in contact with the nucleus is given by the donor occupancy, $F=\Sigma_i P_i$. For instance, if $F=1$, then the nucleus spends negligible time not coupled to any electron. Typically, $F \ll 1$ with the actual value being determined by the intensity of the exciting light. The cross relaxation rate is

$$\frac{1}{T_1^{II}} = \frac{2F^2 a_N^2}{3}\frac{\gamma_e}{\gamma_e^2+\omega_S^2}S(S+1), \tag{12}$$

where $\omega_I - \omega_S \approx -\omega_S$. Finally, the quantity I_0 in Eq. (8) is the expectation value of the nuclear spin at thermodynamic equilibrium (in the dark). It is negligible and will be dropped.

Due to the occurrence of spin diffusion, the polarization of the nuclei can be extended to radii well beyond the Bohr radius a_0. This diffusion sphere is of limited dimension due to the spin lattice relaxation rate T_1 of the bulk nuclei. Addition of the term $\langle I_z\rangle/T_1$ to the left side of Eq. (8) mathematically permits application of the following boundary condition: at sufficiently large r, $\langle I_z\rangle(r,t)=0$ for all t.

To model the dependence of the quantity $\langle I_z\rangle(t,r)$ a modified diffusion equation is constructed by adding spin diffusion and dark relaxation terms to Eq. (8). The result is a second-order, nonlinear partial differential equation in two dimensions that is analogous to the one originally derived by Bloembergen to describe nuclear spin relaxation via diffusion to paramagnetic impurities,[11]

$$\frac{d\langle I_z\rangle}{dt} = D\nabla^2\langle I_z\rangle - \frac{1}{T_1^{II}}\left\{\langle I_z\rangle - \frac{I(I+1)}{S(S+1)}(\langle S_z\rangle - S_0)\right\} - \frac{\langle I_z\rangle}{T_1}. \tag{13}$$

Because GaAs has a cubic lattice, isotropic spin diffusion can be assumed and the term $\nabla^2\langle I_z\rangle$ reduces to

$$\nabla^2\langle I_z\rangle = \frac{1}{r}\frac{\partial^2}{\partial r^2}(r\langle I_z\rangle), \tag{14}$$

independent of ϕ and θ.

The measured quantity in the experiments is the average nuclear polarization following exposure to laser radiation of duration t. This can be calculated from the following formula which sums the expectation values for successive shells of atoms centered at the defect origin weighted by the respective number of nuclei in each shell:

$$\langle I_z\rangle(t) = \frac{n_A}{N}\sum_s \langle I_z\rangle_s (N_s - N_{s-1}). \tag{15}$$

Here, $N_s = 4\pi(2r_s/a)^3/3$ relates the number of atoms contained by the sth shell to the sth shell radius r_s. N is the total number of atoms in the sum, a is the lattice constant for GaAs, and n_A is the natural abundance of the nuclear spin species of interest.

The model used here extends existing theories[3,4] in several ways: (a) the full magnetic field dependence of $\langle I_z\rangle$ is incorporated, (b) the nuclear spin diffusion term $D\nabla^2\langle I_z\rangle$ and ''dark'' relaxation time T_1 have been included, (c) physically meaningful boundary and initial conditions have been employed. These modifications are obviously essential for the correct interpretation of optical pumping signals. In the artificial case where $D=0$, the equation can be trivially integrated, yielding

$$\langle I_z\rangle(t,r) = \frac{I(I+1)}{S(S+1)}(\langle S_z\rangle - S_0)\left(1+\frac{T_1^{II}}{T_1}\right)^{-1} \times\left\{1-\exp\left[-t\left(\frac{1}{T_{II}}+\frac{1}{T_1}\right)\right]\right\}. \tag{16}$$

This expression is useful as a check on the numerical results at $D=0$. In this limit, the steady state value of $\langle I_z\rangle$ is

$$\langle I_z\rangle_\infty(r) = \frac{4}{3}I(I+1)\left[\langle S_z\rangle + \frac{1}{2}\tanh\left(\frac{\hbar g_e\mu_0 B_0}{2kT}\right)\right]\left(1+\frac{T_1^{II}}{T_1}\right)^{-1}. \tag{17}$$

In summary, the spin-diffusion equation has been formulated with the inclusion of the electron-spin thermal polarization factor and the field dependent electron-nuclear spin cross relaxation time constant. The dependence on time, magnetic field, and displacement from the origin are then calculable by numerical resolution of Eq. (13). Using these solutions, the observable is formed using Eq. (15).

IV. EXPERIMENT

The GaAs sample is a semi-insulating (nondoped) substrate single crystal, ingot no. S8870 obtained from Crystal Specialties, Intl. with orientation $\langle 100\rangle 2.0°\langle 110\rangle$, a thickness of 635 μm, and mobility of $(3.1-6.5)\times 10^3$ cm^2/V s.

The optical pumping light was generated by an argon ion pumped continuous wave titanium-sapphire ring system with a maximum output of approximately 1 W. After attenuation the laser beam was focused into a 600 μm diameter fiber. The 20 m fiber terminated at a distance of approximately 0.75 cm from the surface of the crystal, yielding a laser spot size of 0.3 cm with a transmission efficiency of 75%. Light transmitted by the fiber emerges unpolarized. At this power level the available wavelength range from the Ti:sapphire system was typically 790−910 nm. In measuring the wavelength dependence of the optical pumping signal the power was maintained at a constant level (to within a few percent) using a variable neutral density filter.

To measure the efficiency of the optical pumping over the magnetic field range of 0−24 T, it was necessary to employ the resistive Bitter magnets at NHMFL. These magnets are capable of ramping at the rate of 0.6 T/s. The FWHM line width of the optically pumped ^{69}Ga resonance at a Larmor frequency of 150 MHz was measured to be $\approx$17 ppm. Spatial field homogeneity over the dimensions of the 4 mm$\times$4 mm sample and temporal stability were both smaller than this value.

The NMR probe consisted of a bottom tuned circuit prematched to compensate for low temperature. Low temperature Q factors of up to 500 were typically obtained. In working with the resistive magnets, precise tuning of the NMR rf circuit is not necessary because the magnetic field

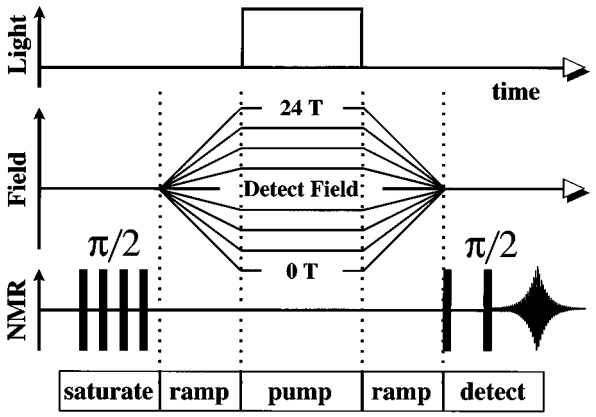

FIG. 1. Timing diagram for obtaining the field dependence of the optical Overhauser effect. The experiment begins with saturation of the nuclear polarization with a train of $\pi/2$ rf pulses. The probe tuning is fixed for resonance at the initial value of the magnetic field. The magnetic field of the resistive Bitter type magnet is ramped at a rate of 0.3 to a specified ''pumping'' field, in the range of $0-24$ T. After reaching the target field, the sample is exposed to laser light for a duration of $15-60$ s following which the magnetic field is returned to the initial value. The free induction decay is acquired using a solid echo sequence, $(\pi/2_x-\tau-\pi/2_y-\tau-\text{acq.})$. The field ramps were repeated twice, once with irradiation and once without, so that the repolarization due to dark relaxation can be accounted for.

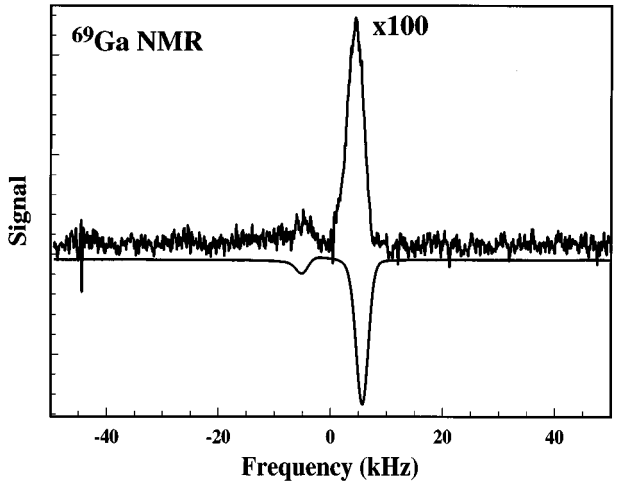

FIG. 2. A representative ^{69}Ga NMR spectra of semi-insulating, bulk GaAs, obtained with (bottom trace) and without enhancement by the optical Overhauser effect. The dark signal has been vertically scaled by a factor of 100. Both spectra were processed and phased identically. The light signal was obtained after a 60 s exposure at $\lambda_{ex}=825$ nm at a field of 4.1 T with NMR detection at 14.88 T. The small peak at -10 kHz is a ''quad ghost.''

can be easily moved around to obtain resonance. Experiments were performed at either 4.2 K or 1.5 K. The sample was always in direct contact with the liquid helium.

The field dependence was obtained using a field cycling procedure whereby the NMR detection frequency was held constant. This is made possible by the long spin lattice relaxation time (tens of minutes) in undoped GaAs at temperatures of a few kelvin. This field cycling procedure has the advantage of not requiring probe tuning each time the pumping field is changed. Consequently, the frequency dependence of the spectrometer detection efficiency does not need to be known as it would be were the probe retuned to resonance for each value of the magnetic field. The minimal relaxation occurring during the ramping or in the ''bulk'' part of the sample can be subtracted away and does not introduce significant errors in the measurements. As shown in the timing sequence of Fig. 1, the experiment begins with saturation of the nuclear spin resonance of ^{69}Ga by a train of resonant $\pi/2$ pulses. The field is then ramped at a rate of 0.3 T/s to a set optical pumping field. When the target field has been reached, the sample is exposed to the light for a duration of $15-60$ s. The sample is subsequently returned to the NMR detection field where the longitudinal magnetization of the ^{69}Ga is sampled in one shot using a $(\pi/2_x-\tau-\pi/2_y-\tau-\text{acq.})$ solid echo pulse sequence.[12] The *saturate-ramp-pump-ramp-detect* cycle is repeated for the entire series of magnetic fields from $0-20$ T or $0-24$ T. The nearly negligible dark signals were acquired for every other value of the field with values in between generated by interpolation. The dark signals were subtracted from the signals obtained with the light on.

To eliminate the possibility that the field dependence is influenced by the choice of the detection field, the field dependence of optical pumping in GaAs was repeated at two different detection fields: 4.97 T and 14.89 T.

V. DISCUSSION

A typical optically pumped NMR spectrum is shown in Fig. 2 along with the ''dark'' signals obtained under identical conditions except without exposure to laser light. The enhanced ^{69}Ga line shapes are Gaussian, characterized by a field independent FWHM of 2.5 kHz. The signal enhancement with unpolarized light is unambiguously 180° out of phase with respect to the dark signal. As is immediately evident from Eq. (8), opposing phases of the optical pumping and dark signals are expected when the signs of the g factor and nuclear gyromagnetic ratio are opposite. For GaAs the literature conduction electron value is $g_e=-0.44$ and the gyromagnetic ratio of ^{69}Ga is positive in sign.[13] This is valid because the g factor of shallow bound donors in GaAs is known from optically detected ESR experiments to be negligibly different from the value for conduction electrons.

Shown in Fig. 3 is the dependence of the ^{69}Ga optically pumped NMR signal integral on the optical excitation energy at 2, 7, 15 and 20 T. The enhancement is peaked at an energy slightly below the free exciton band gap of 1.519 eV with a width of ≈ 20 meV with the maximum shifting to higher energy by approximately the same amount over the $2-20$ T range. Since a single optical excitation energy is used in the field dependent studies, the shift of the maximum in the optical pumping excitation energy response must be considered. This influence was noted when the GaAs field dependence was repeated for two different values of the excitation energy, as indicated by ''*a*'' and ''*b*'' on the figure. For case *a*, the field dependence was more strongly affected as the peak is swept through the excitation wavelength. In case *b* the influence of the shift is still present but smaller.

The magnetic field dependence of the optical pumping signals is shown in Fig. 4. The general appearance of the

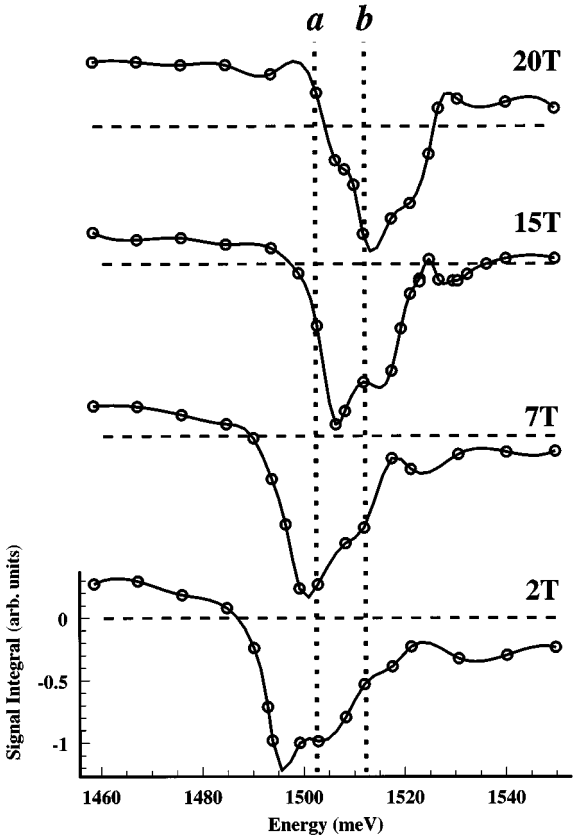

FIG. 3. Optical excitation energy dependence of the optical Overhauser NMR signal enhancement of ^{69}Ga in GaAs, recorded at 2, 7, 15, and 20 T using the field ramp method (see Fig. 1). The field dependence (Fig. 4) was obtained at two different values of the excitation energy, position ''*a*'' with $\lambda_{ex}=825$ nm and position ''*b*'' with $\lambda_{ex}=820$ nm. The NMR was detected at a field of 7 T.

curves, in which the signal sharply increases with magnetic field, passes through a maximum and then monotonically approaches zero with increasing field, reflecting in qualitative terms the competition between the increasing deviation of the electron spin polarization from Boltzmann equilibrium versus the monotonically decreasing cross relaxation rate T_1^{II}. A quantitative model for the field dependence is obtained by solving Eq. (13). This is achieved by imposing a boundary condition whereby the slope $|dI_z(r,t)/dr|_{r=0}$ vanishes for all t. This is realistic because of the discrete spacing of nuclei in the lattice. If $t=0$ denotes the moment at which the sample is first exposed to light, then a second boundary condition is given by $\langle I_z\rangle(t\rightarrow 0,r)=0$ for all r given that the sample is initially unpolarized.

Numerical solutions of Eq. (13) were obtained using PDECOL,[14] a general collocation algorithm for partial differential equations. The boundary conditions are specified in the syntax of PDECOL as follows.

At $r=0$, $B=d\langle I_z\rangle/dr$ for all t,

$$\frac{dB}{d\langle I_z\rangle}=0, \quad \frac{dB}{d\langle I_z\rangle/dr}=1, \quad \frac{dz}{dt}=0. \tag{18}$$

At $r=100a_0$, $B=\langle I_z\rangle$ for all t,

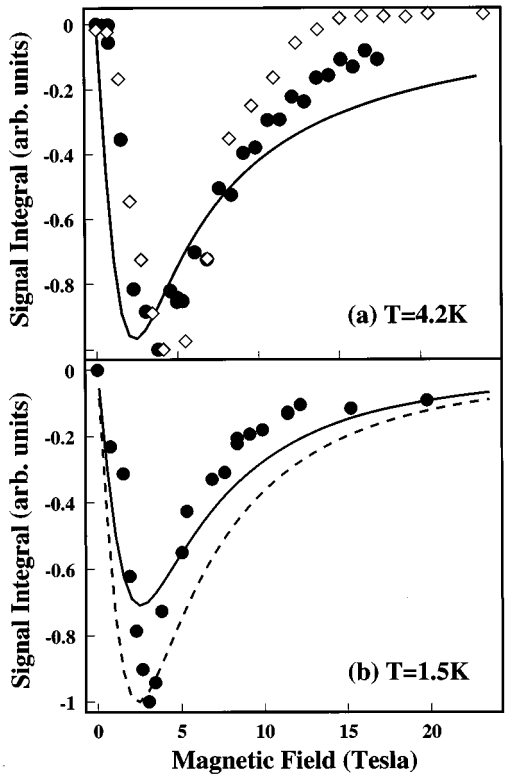

FIG. 4. Field dependence of the optical Overhauser enhancement. (a) $T=4.2$ K. The ^{69}Ga NMR signals were recorded following optical pumping with unpolarized light and under two sets of conditions. (i) with $\lambda_{ex}=825$ nm detected at a field of 14.89 T (diamonds). (ii) With $\lambda_{ex}=820$ nm (filled circles) and detected at 4.97 T. The solid line represents the simplex least squares fit of the diffusion equation to the experimental data obtained under condition (i). (b) $T=1.5$ K. Signals obtained with $\lambda_{ex}=820$ nm (filled circles) and detected at 4.97 T. The solid line represents the three parameter simplex fit. The dashed line was generated using the fitted parameters from the $T=4.2$ data shown in part (a) but at $T=1.5$ K. All of the field dependence curves have been normalized.

$$\frac{dB}{d\langle I_z\rangle}=1, \quad \frac{dB}{d\langle I_z\rangle/dr}=0, \quad \frac{dz}{dt}=0. \tag{19}$$

Using these boundary conditions, the theoretical optically pumped NMR signal intensity is calculated from numerical solution of Eq. (8). These solutions, along with Eq. (15), are used to compute the observable. Least squares fits of the experimental data to the calculated value were obtained by Simplex minimization. The data and fits are presented in Fig. 4. Table I summarizes the parameter values yielded by this procedure. To check that $r=100a_0$ is a good approximation for the infinite radius boundary condition, the $\langle I_z\rangle(r)$ obtained using Eq. (15) were also calculated using $r=50a_0$ and found to be negligibly different.

The fitted parameters were generated by fixing g_e to the literature value obtained by optically detected ESR. As mentioned earlier, a negative g_e is consistent with the observed field dependence in GaAs. The average hyperfine frequency, $|Fa_N|$, should depend on the intensity of the optical field. The nuclear spin diffusion constant has been measured previously for ^{75}As nuclei in GaAs. This value, along with the approximate relation[15] $D=\gamma_n\Delta\nu d^2/30$, where $\Delta\nu$ is the

TABLE I. GaAs fitted parameters.

Method	T (K)	γ_e (s^{-1})	$\lvert Fa_N\rvert$ (s^{-1})
GaAs, fixed $g_e=-0.44$ [a]	4.2	7.2×10^{10}	1.1×10^{6}
	1.5	8.9×10^{10}	0.76×10^{6}
Literature and estimate[b,c]		$\approx5.0\times10^{10}$	

[a]Reference 13.
[b]Reference 3.
[c]Reference 4.

FWHM of the NMR transition, γ_n is the nuclear gyromagnetic ratio, and d is the internuclear spacing, can be used to estimate the value for ^{69}Ga. A value of $D=3000$ Å^2/s was used. The correlation frequency γ_e that emerges from the fits agrees well with the estimated value in the literature.

The inability to obtain exact correspondence between the theoretical model and the data may be hampered by several factors, as follows. (i) Due to the field shift of the sharp maximum in the optical pumping wavelength dependence in GaAs, distortion of the field dependence occurs as the laser wavelength is swept through this peak. (ii) The correlation function chosen to describe the hyperfine fluctuations may have a more complex form or may have some intrinsic field dependence. (iii) The coefficient of nuclear spin diffusion is likely to possess some (weak) field dependence that has not been taken into account. (iv) The data have been corrected for dark relaxation by subtracting the signals obtained with and without laser irradiation. This assumes that the background relaxation is the same whether the light is on or off. However, this assumption might not be valid if paramagnetic deep trap states are generated by the light. This type of relaxation would have its own field dependence. (v) The low field dependence of $\langle S_z\rangle$, which varies according to Eq. (3), has not been included in Eq. (13). Consequently, the field dependence below $B_0\approx0.5$ T is not expected to be properly modeled. This will have a lesser impact in the case of unpolarized light since the electron spin polarization vanishes at zero field anyway. The fits should not be significantly affected since only a few data points were recorded at these low fields. On the other hand, the predictions for circularly polarized light are expected to deviate strongly with experimental data at very low field. However, given the limitations outlined above, the model accounts for the overall optical pumping field dependence satisfactorily over the entire range of magnetic fields.

Having obtained values for the three fitted parameters, γ_e, D, and $F^2a_N^2$ it is now possible to use Eq. (13) to predict the field dependence of the signal using circularly polarized light. In this case, σ^+ light yields $\langle S_z\rangle=+1/4$ and σ^- light produces $\langle S_z\rangle=-1/4$. The curves for GaAs are shown together with the data and fits for $\langle S_z\rangle=0$ in Fig. 5(a). Note that the optical pumping efficiency is greatly diminished above about 20 T. It is also apparent that pumping with σ^- light produces maximal nuclear polarization at all values of the field. The curves corresponding to other g factors, holding all other parameters constant, can be predicted. For example, the curves for g factors of $+2.0$, -0.1, -0.44, and -5.0 are presented in Fig. 5(b).

VI. CONCLUSIONS

To summarize, the field dependence of the optical Overhauser effect has been measured in semi-insulating GaAs crystals. The phase of the optically enhanced signal obtained with unpolarized light is consistent with scalar relaxation with $g_e<0$. The theoretical field dependence is based on scalar relaxation of the first kind between the weakly bound donor electrons and the nuclei within their Bohr radii. The inclusion of spin diffusion and a background ''dark'' relaxation time limits the magnitude of the steady state nuclear magnetization that is obtained. The numerical reso-

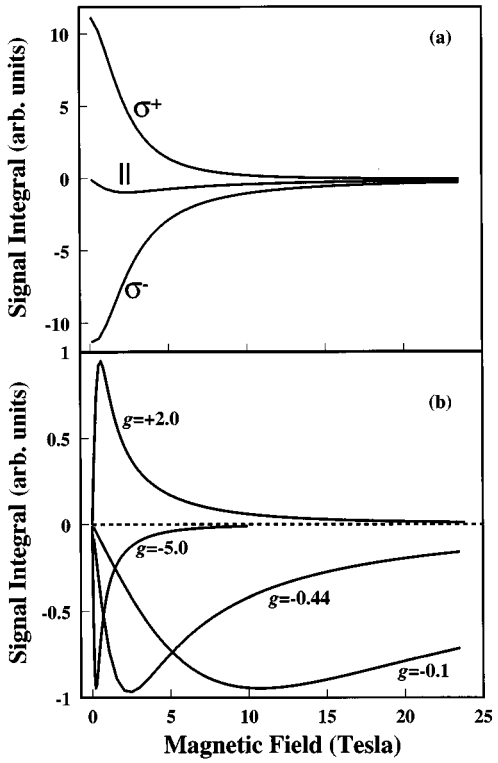

FIG. 5. (a) Theoretically predicted field dependence of optically enhanced NMR signals for GaAs at 4.2 K for σ^+, σ^- and unpolarized laser light. Also shown are the fits to the data obtained using unpolarized ($\|$) light. The theoretical plots are based on the fitted values for the parameters γ_e and $\lvert Fa_N\rvert$ given in Table I and the fixed parameters $g_e=-0.44$ and $D=3000$ obtained from the literature. (b) Hypothetical optical Overhauser effect field dependence curves for various g factors generated by fixing all other parameters to the Table I values. At higher g_e, the maximum pumping effect shifts to lower field and the field dependence becomes more peaked in shape.

lution of the diffusion equation yielded the radial dependence of $\langle I_z \rangle (r,t)$, which was then summed over sequential atomic shells to give the observed quantity $\langle I_z \rangle$. For unpolarized light, the magnitude of $\langle I_z \rangle$ increases from zero at zero field, maximizes at a field of $2-4$ T, then monotonically decreases to zero at very high field.

The value of the field that gives optimal optical pumping with unpolarized light is determined primarily by the magnitude of the g factor and the correlation time $1/\gamma_e$. The two parameter fits yielded values for the electron spin correlation time and the average hyperfine field. The values are in satisfactory agreement with literature values. This instills confidence that the model postulated herein correctly describes the true dynamics of optical pumping in semi-insulating GaAs.

Finally, it has been shown that optical pumping is quenched at fields above about 20 T and that the efficiency of optical pumping using either circular or unpolarized light depends strongly on the magnitude of the g factor. This ultimately determines the maximum field at which optical pumping can be employed.

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