

# Optical polarization of nuclei in GaAs/AlGaAs quantum-well structures

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A polarization of the nuclei of the crystal lattice by optically oriented electrons has been detected in quantum-well structures for the first time. An expression is derived for the rate of nuclear polarization by a nondegenerate 2D electron gas. It is concluded from a comparison of the theoretical and experimental longitudinal relaxation times of nuclei that in the temperature range 2–77 K the localized charge carriers are responsible for the polarization of nuclei in the GaAs/AlGaAs quantum wells which were studied.

Optical orientation has been widely used over the past two decades to polarize and study the properties of the nuclear spin systems of bulk semiconductors.<sup>1–4</sup>

In this letter we are reporting the first observation of a polarization of the nuclei of the crystal lattice by optically oriented carriers in quantum-well structures. The nuclear polarization achieved in GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum wells over the temperature range 2–77 K was a few percent. This significant polarization is evidence of a localization of the charge carriers responsible for the polarization of the nuclei. To prove that this is the case, we derive an expression for the time scale of the longitudinal nuclear relaxation resulting from the interaction with a 2D electron gas. The value estimated for this time is much longer than the time observed experimentally, supporting our conclusion about the role of localized states.

1. The experiments were carried out on a GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum-well heterostructure<sup>5</sup> with a single quantum well 100 Å thick over the temperature range 2–77

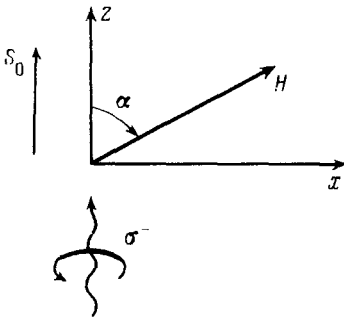


FIG. 1. Experimental geometry.

K. The structures were grown by metalorganic hydride epitaxy. Optically polarized 2D electrons in the GaAs layer were excited by  $\sigma$ -polarized light with a wavelength of 7525 Å, which propagated along the  $z$  axis, normal to the surface of the sample (Fig. 1). The measured quantity was  $\rho$ , the degree of circular polarization of the luminescence detected in a reflection arrangement along the  $z$  axis. Here  $\rho \propto S_z$ , where  $\vec{S}$  is the average spin of the electrons.

Measurements of curves of the magnetic depolarization of the luminescence make it possible to study the optical polarization of the nuclei. The reason is that the optical orientation of the electrons in semiconductors of the GaAs type is accompanied by a dynamic polarization of the nuclei of the crystal lattice.<sup>1-4</sup> In turn, the effective field of the polarized nuclei,  $\vec{H}_N$ , acts on the average spin of the oriented electrons,  $\vec{S}$ , giving rise to a complex dependence of the luminescence polarization on the external magnetic field<sup>2-4</sup>  $\vec{H}$ .

The nuclear field  $\vec{H}_N$  is manifested most commonly when  $\vec{H}$  makes an angle  $\alpha$  with respect to the exciting light (Fig. 1). In this case the field  $\vec{H}_N$  either intensifies or

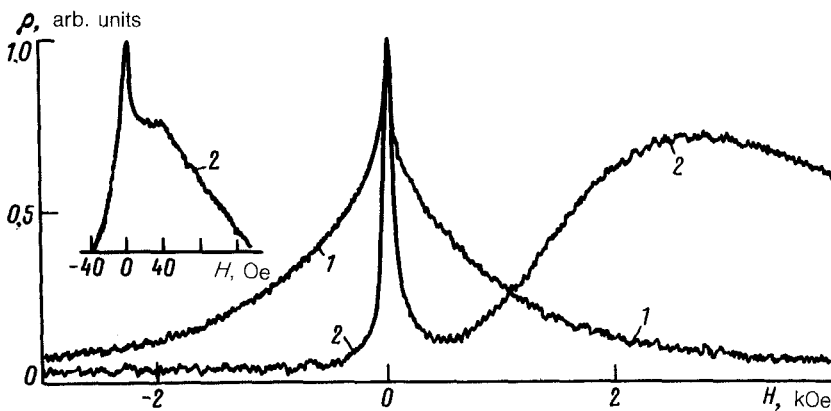


FIG. 2. Experimental Hanle curves for an isolated quantum well. 1—The circular polarization of the exciting light is varying at a frequency of 34 kHz; 2—this polarization is constant ( $T = 2$  K,  $\alpha = 85^\circ$ ).

weakens (depending on its sign) the effect of the component of the external field which is transverse with respect to  $\vec{S}$ .

Figure 2 shows experimental curves of the magnetic depolarization of the luminescence of the test structure for an angle  $\alpha = 85^\circ$  at  $T = 2$  K.

In an effort to avoid nuclear effects,<sup>2</sup> we measured curve 1 as the circular polarization of the exciting light was varied at a high frequency. For this purpose, we passed the exciting beam through a photoelastic polarization modulator<sup>7</sup> operating at a frequency of 34 kHz. In this case the dynamic polarization cannot keep up with the rapidly changing direction of the average electron spin, and there is no nuclear field  $\vec{H}_N$ . Curve 1 is thus a purely electronic Hanle curve, symmetric with respect to the ordinate, with a maximum at  $H = 0$ .

Curve 2 was recorded at a constant circular polarization of the exciting light. We see two additional maxima: at  $H \approx 2.5$  kOe and  $H \approx 40$  Oe. The maximum at a high magnetic field stems from a cancellation of the external field by the nuclear field  $H_N \propto S_H = S(H=0)\cos\alpha$  (Refs. 2 and 3). The additional maximum at a comparatively weak field shown in the inset in Fig. 2 arises from a cancellation of the longitudinal component of the external field,  $H_Z$ , by the hyperfine-interaction field  $H_e$ , caused by the polarized electrons at nuclei.<sup>3</sup> Near this maximum, a nuclear polarization does not arise, since in this case the nuclei are in a resultant magnetic field with an absolute value close to zero. The direction of the nuclear field,  $\vec{H}_N$ , is determined by the sign of the product of the nuclear gyromagnetic ratio  $\gamma$  and the electron  $g$ -factor. In the structures which were studied, the relation  $\gamma > 0$  holds for all isotopes. It turns out<sup>3</sup> that in crystals with a negative  $g$ -factor the additional maxima correspond to the same sign of the external field  $H$ , while in the case  $g > 0$  they correspond to different signs. It can be seen from curve 2 in Fig. 2 that the electron  $g$ -factor in the quantum well is negative, as in bulk GaAs.

Direct confirmation of a polarization of nuclei comes from observation of the NMR signal. During the application of an rf field at the NMR frequency, the polarization of the nuclei decreases, so the field  $H_N$  also decreases. This decrease of the nuclear field causes a sharp change in  $\rho$  if the value of  $H$  near the additional maximum, at which the nuclear field is canceled by the external field, is chosen in such a way that the derivative  $d\rho/dH$  is large. Figure 3 shows an NMR spectrum of the GaAs quantum-well layer recorded as the frequency of the rf field, with an amplitude  $2H_1 = 0.48$  Oe, was varied slowly in a field  $H = 753$  Oe. It can be seen from this figure that signals were detected from all three isotopes of the GaAs crystal lattice.

Measurements of the nuclear field  $H_N$  by means of the Hanle effect in an oblique field make it possible to determine the average nuclear spin,  $\langle I \rangle$ , since we have  $\vec{H}_N = h_N \langle I \rangle$ . From curve 2 in Fig. 2 we find  $H_N \approx 2.5$  kOe. Using the value<sup>8</sup>  $h_N = 35.3$  kOe calculated for bulk GaAs, we find  $\langle I \rangle \approx 7\%$  in the given sample. Up to  $T = 77$  K, the nuclear polarization is  $\sim 1\%$ .

The longitudinal relaxation time  $T_1$  of the nuclei was determined from the transients upon the application of an rf field at the NMR frequency. The result is  $T_1 < 1$  s.

2. To determine which electrons are responsible for the polarization of the nuclei in the quantum wells, let us estimate the time scales of the nuclear relaxation due to

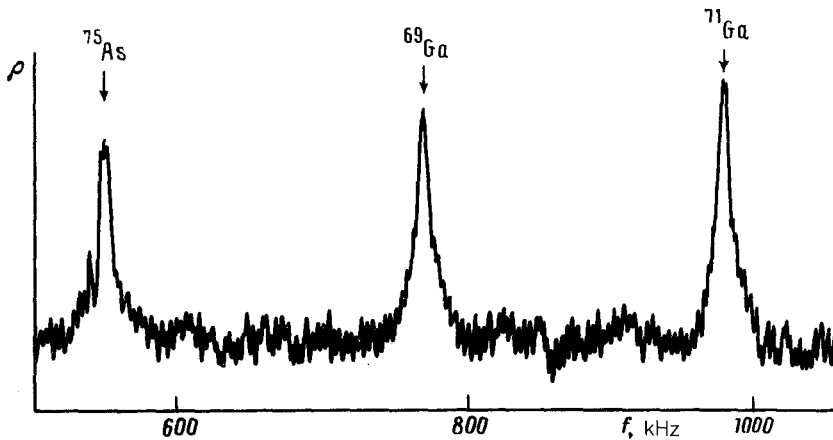


FIG. 3. Nuclear-magnetic-resonance spectrum of a GaAs quantum-well layer 100 Å thick ( $H = 753$  Oe,  $\alpha = 85^\circ$ ,  $T = 2$  K). The amplitude of the rf field is  $2H_1 = 0.48$  Oe.

the contact hyperfine interaction with free and localized electrons and then compare these times with the time  $T_1$  found experimentally.

For this estimate we use the customary dynamic-averaging formula

$$T_{1e}^{-1} = \omega^2 \tau_C,$$

where  $\omega$  is the precession frequency of the nuclear spin in a random hyperfine-interaction field, and  $\tau_C$  is the correlation time of this field. If an electron occupies a volume  $v_0$ , we can find  $\omega^2$  from<sup>4</sup>

$$\omega^2 \propto (A/\hbar N)^2 F.$$

Here  $A$  is the hyperfine constant,  $N \sim v_0/\Omega$  is the number of nuclei in a volume  $v_0$  ( $\Omega$  is the volume of the unit cell), and  $F$  is the number of electrons in this volume. The quantity  $A/(\hbar N)$  is the precession frequency of the nuclear spin in the magnetic field produced by one electron.

For nondegenerate free electrons in a quantum well of width  $d$ , the volume  $v_0$  is on the order of  $\lambda^2 d$ , where  $\lambda$  is the de Broglie wavelength (in a bulk semiconductor we would have  $v_0 \sim \lambda^3$ ). In this case we have  $F \propto n v_0$  ( $n$  is the density of electrons), and the correlation time is the time required for an electron to traverse a distance on the order of  $\lambda$ :  $\tau_C \sim m \lambda^2 / \hbar$ , where  $m$  is the effective mass of the electrons. We thus have

$$T_{1e}^{-1} \propto A^2 \Omega^2 n m \hbar^{-3} d^{-1}. \quad (1)$$

A rigorous calculation carried out by the standard procedure,<sup>9</sup> under the assumption of an infinitely high barrier and a freezing of the free electrons at the first level in the quantum well, shows that the rate of the spin-lattice relaxation of the nuclei at a distance  $z$  from the center of the well is

$$T_{1e}^{-1}(z) = A^2 \Omega^2 n m \hbar^{-3} d^{-1} \cos^4(\pi z d^{-1}). \quad (2)$$

With  $d = 100 \text{ \AA}$ ,  $n = 10^{15} \text{ cm}^{-3}$ , and  $A = 10^{-16} \text{ erg}$ , we find a time  $T_{1e}(0) = 500 \text{ s}$  for the nuclei at the center of the well.

Significantly, the time scale of the nuclear relaxation involving the free electrons in a quantum well does not depend on the temperature, in contrast with the case of a bulk semiconductor, in which we would have<sup>9</sup>  $T_{1e} \propto T^{-1/2}$ .

We can use expression (1) to estimate the rate of the relaxation involving free excitons. As  $n$  and  $m$  in (1) in this case we should use the density and translational mass of the excitons. It is not difficult to see that the time scale of the nuclear relaxation involving free excitons is on the same order of magnitude as that involving free electrons.

For electrons localized at donors in a narrow well of thickness  $d \ll a_B$  ( $a_B$  is the first Bohr radius of the electron), we have  $v_0 \sim a_B^2 d$ . In this case  $F = n/N_D$  is the extent of donor filling ( $n$  and  $N_D$  are the densities of bound electrons and of donor centers), and  $\tau_C$  is the time an electron spends at a donor. For nuclei in the region within which an electron is localized at a donor we then have

$$\frac{1}{T_{1e}^D} \propto \frac{A^2 \Omega^2}{\hbar^2 a_B^4 d^2} \frac{n}{N_D} \tau_C. \quad (3)$$

For the structures studied we have<sup>5</sup>  $N_D \approx 10^{16} \text{ cm}^{-3}$ . The electron lifetime determined from the half-width of the Hanle curve (curve 1 in Fig. 2) is  $\tau \approx 10^{-9} \text{ s}$ . At the light intensity used, we then find  $n \sim 10^{15} \text{ cm}^{-3}$  and  $n/N_D \sim 0.1$ . The time  $\tau_C$  is not known accurately. It is the shortest of the times determined by recombination, a hopping conductivity, thermal ionization, and the spin relaxation of electrons. The time  $\tau \approx 10^{-9} \text{ s}$  is evidently an upper limit on  $\tau_C$ . At liquid-helium temperatures, the hopping conductivity and the spin exchange with free electrons are the fastest processes; the respective time scales are  $10^{-11}$ – $10^{-10} \text{ s}$  (Ref. 10) and  $\sim 10^{-11} \text{ s}$  (Ref. 11). In this case, with  $d = a_B = 100 \text{ \AA}$ , we find  $T_{1e}^D \sim 0.1$ – $0.01 \text{ s}$ . With increasing temperature, thermal ionization may become more effective, but at  $\tau_C > 10^{-12} \text{ s}$  we have  $T_{1e}^D < 1 \text{ s}$ .

These estimates indicate that the rate of polarization of the nuclei in quantum wells by localized electrons is significantly higher than that of the polarization by free electrons or excitons. A comparison of the calculated times with the time  $T_1 < 1 \text{ s}$  found experimentally permits the conclusion that localized electrons and excitons are responsible for the polarization of the nuclei in the quantum wells studied in the temperature range  $T = 2$ – $77 \text{ K}$ .

In summary, this study has yielded the first observation of an optical polarization of the nuclei of the crystal lattice of GaAs quantum-well layers. A study of this polarization in an oblique magnetic field has revealed the sign of the electron g-factor and the time scale of the spin-lattice relaxation of the nuclei. An expression has been derived for the rate of spin-lattice nuclear relaxation due to the interaction of the nuclei with a nondegenerate 2D electron gas. It has been shown that the polarization

of the nuclei in the quantum wells studied is due primarily to localized electrons and excitons.

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