Electrical Detection of Nuclear Magnetic Resonance in GaAs-$Al_xGa_{1-x}As$ Heterostructures

M. Dobers and K. v. Klitzing
Max-Planck-Institut für Festkörperforschung, Heisenbergstrass 1, D-7000 Stuttgart 80, Federal Republic of Germany

J. Schneider
Fraunhofer Institut für Angewandte Festkörperphysik, Eckerstrasse 4, D-7800 Freiburg, Federal Republic of Germany

G. Weimann
Walter-Schottky-Institut, Technische Universität München, D-8046 Garching, Federal Republic of Germany

and

K. Ploog
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 27 June 1988)

The experimental investigation of the electron spin resonance in the two-dimensional electron gas of GaAs-$Al_xGa_{1-x}As$ heterostructures via the ESR-induced change of magnetoresistivity reveals hysteresis and long-persisting memory effects. We have been able to prove the nuclear origin of the observed effects by performing nuclear magnetic resonance and observing NMR in the magnetoresistivity of the heterostructures.

PACS numbers: 72.20.My, 72.80.Ey, 76.30.Pk, 76.70.Fz

The spin splitting of the Landau levels in the two-dimensional electron gas of a GaAs-$Al_xGa_{1-x}As$ heterostructure can be determined by ESR. For this, conventional ESR techniques are not sensitive enough as a result of the small number of electrons in the 2D electron gas. However, the magnetoresistivity ρ_{xx} is affected by ESR[1] and the device itself can be used as a sensitive detector. The measured spin splitting is not solely due to the Zeeman effect, but is also influenced by the nuclear-spin polarization, which acts via the hyperfine interaction on the electronic spins[2,3]: The Hamiltonian of an electronic spin $\mathbf{S}$ interacting with a nuclear spin $\mathbf{I}$ is thus the following:

$$H = g\mu_B \mathbf{B}\cdot\mathbf{S} + A\mathbf{I}\cdot\mathbf{S}, \tag{1}$$

with g being the effective electronic g factor, μ_B the Bohr magneton, $\mathbf{B}$ the magnetic field, and A the hyperfine interaction constant. Averaging over all nuclear spins leads to the electronic spin splitting

$$\Delta E = +g\mu_B B + \tfrac{8}{3}\pi g_0 \mu_B g_N \mu_N |\psi(0)|^2 \langle I\rangle, \tag{2}$$

where g_0 is the free-electron g factor, g_N and μ_N are the nuclear g factor and magneton, respectively, $|\psi(0)|^2$ is the probability of finding the electron at the nuclear site, and $\langle I\rangle$ is the nuclear-spin polarization. In thermodynamic equilibrium $\langle I\rangle$ is negligible even at liquid-helium temperatures. Thus the electronic spin splitting is, in this case, exclusively determined by the first term, the Zeeman term. By performing ESR, however, the nuclear spins may be dynamically polarized[2,3] and hence the second term, the hyperfine term, becomes important. One possible channel for electronic spin relaxation is the combined flip-flop of an electronic and nuclear spin, which is brought about again by the hyperfine interaction,

$$A\mathbf{I}\cdot\mathbf{S} = A[0.5(I_+S_- + I_-S_+) + I_zS_z]. \tag{3}$$

This dynamical nuclear-spin polarization results in a shift of the ESR line, the Overhauser shift B_N,[2,4] which from Eq. (2) can easily by calculated:

$$\begin{aligned}\Delta E &= g\mu_B[B + B_N] \\ &= g\mu_B[B + \tfrac{8}{3}\pi g_0 g_N \mu_N |\psi(0)|^2 \langle I\rangle/g].\end{aligned} \tag{4}$$

Up to now this effect has been seen in metals,[2,5] bulk semiconductors,[6] and organic materials.[7] In this Letter we report the first observation of the Overhauser shift in a two-dimensional electron system. By applying radio frequencies to saturate the NMR we have been able to prove the nuclear origin of the observed shifts and to detect NMR in the magnetoresistivity.

All heterostructures showing ESR revealed the Overhauser shift, but the results from only one device are reported here as an example. The sample is a single-side-doped quantum well with a well width of 15 nm GaAs, a two-dimensional carrier density of $N_s = 3.3 \times 10^{11}$ cm^{-2}, and a mobility of 300000 cm^2/V s at 4.2 K (sample 2 in Ref. 1). The magnetoresistivity ρ_{xx} was measured at liquid-helium temperatures in magnetic fields up to 14.5 T under microwave radiation in the frequency range up to 70 GHz with an output power between a few mW and several hundred mW.

Figure 1 shows traces of ρ_{xx} under microwave radiation. At $B=5.5$ T ESR occurs and manifests itself in a strong change of ρ_{xx} yielding a sharp resonance structure. This sharp ESR structure visible in the upward sweep of the magnetic field, $\dot{B}>0$ (solid line), disappears for a downward magnetic field sweep, $\dot{B}<0$ (dashed line), where the resonant structure is largely smeared out. This and all other experimental data agree with the picture that the absorption process during ESR shifts the resonance to lower magnetic fields.

This hysteretic behavior is due to the dynamic polarization of the nuclear spins. The ESR-induced nuclear polarization produces an equivalent nuclear magnetic field B_N, which shifts the resonance position $B_{\rm res}$ towards lower fields: $B_{\rm res}=B^0_{\rm res}-B_N$, $B^0_{\rm res}$ being the unshifted resonance position. Thus an increasing $\langle I\rangle$ leads to a decreasing resonant magnetic field: $\dot{B}_{\rm res}<0$. If the magnetic field is swept up, a crossing of the applied external field, $\dot{B}>0$, with the resonant magnetic field, $\dot{B}_{\rm res}<0$, occurs. Because of the opposite movement of these two fields the resonance condition is fulfilled only within a small magnetic field range, of the order of the ESR linewidth (≈ 30 mT). The nuclear-spin polarization remains low, since the ESR condition is fulfilled only for a short time. On the other hand, if one decreases the external magnetic field, both this and the resonance field are decreased: $\dot{B}<0$, $\dot{B}_{\rm res}<0$. As a result of this parallel movement of the two fields, the resonance condition is fulfilled during a much longer time, so that the nuclear-spin polarization is increased steadily. At microwave powers of the order of 100 mW at the source, the shift of the resonance position under ESR conditions was faster than the sweep rate of the external magnetic field ($\dot{B}=-2.35$ T/min), so that a crossing was not possible; instead, the ESR position was continually pushed just below the external magnetic field value. On stopping the magnetic field sweep, the resonance shifts further on to lower fields until the electronic spins are out of resonance, and $\langle I\rangle$ cannot be increased any longer. Thus, in this case, the resonance position after stopping a downward sweep is just below the applied external magnetic field.

A nonequilibrium nuclear-spin polarization, once created, is, on the time scale of our experiments, approximately stable. This is illustrated in Fig. 2. The lower trace is ρ_{xx} without microwaves and the three upper traces are measurements of ρ_{xx} with microwaves. The first measurement with microwaves is the dash-dotted trace. The sample, being in thermal equilibrium ($\langle I\rangle\approx 0$), shows ESR at $B\approx 5.5$ T, sweeping up. The next measurement, the dashed trace, is taken sweeping down. ESR occurs at the same magnetic field as in the sweep-up measurement. However, by polarizing the nuclear spins the resonance is pulled towards lower magnetic fields. During the uninterrupted down-sweep, at $B\approx 5.3$ T the microwaves have been switched off and, immediately, ρ_{xx} falls down to its nonradiated value.

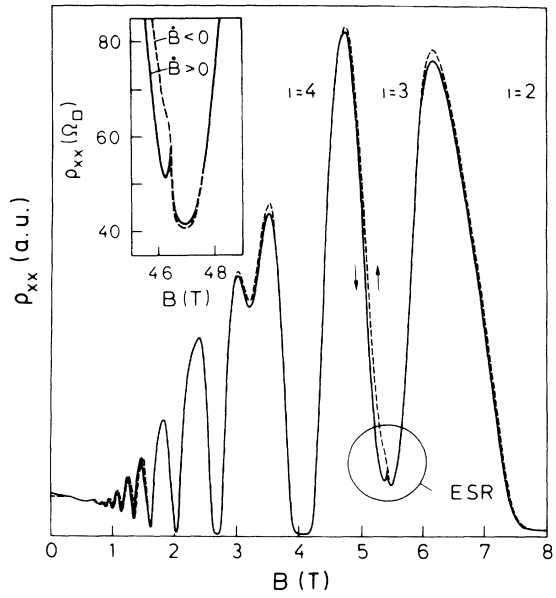

FIG. 1. Magnetoresistivity ρ_{xx} under microwave radiation with a frequency $\nu=23.1$ GHz as a function of the magnetic field. Solid line, $\dot{B}=+2.35$ T/min; dashed line, $\dot{B}=-2.35$ T/min. The filling factor i for some of the minima is indicated. The sharp structure at $B=5.5$ T is due to the conduction-electron-spin resonance. Similar traces of ρ_{xx} in the vicinity of $i=3$ are shown in the inset on a larger scale. The sample normals are tilted by angles in the order of 30° with respect to the magnetic field.

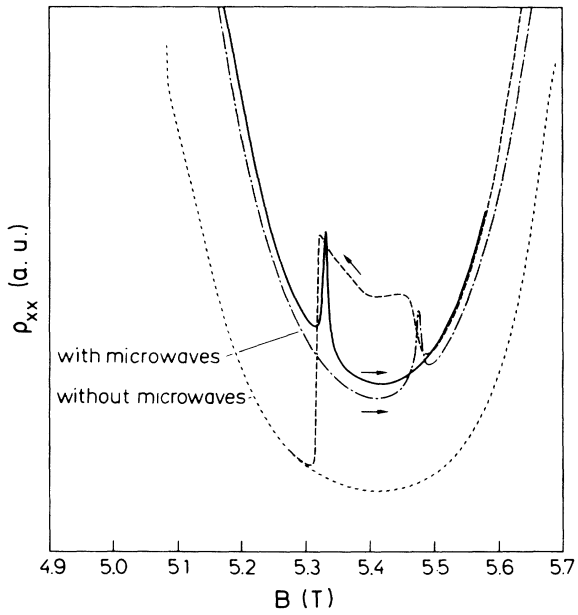

FIG. 2. Magnetoresistivity ρ_{xx} in the vicinity of filling factor $i=3$ without (lower trace) and with microwave radiation; $\dot{B}=\pm 2.35$ T/min, the sign of $\dot{B}$ being indicated by the arrows.

Without ESR the nuclear-spin polarization is not increased further, but remains approximately constant, so that the equivalent nuclear magnetic field, in this case $B_N \approx 0.2$ T, also remains constant. The following measurements with microwaves again switched on and $\dot{B} > 0$, solid line, reveals ESR still at the shifted position, where the microwaves had been switched off. The memory for this shift, i.e., the nuclear-spin relaxation time T_1 can be determined by studying the backshift of the ESR line. T_1 depends on the temperature and the magnetic field and is typically in the order of 20 min in the experiments shown in this Letter. Obviously, T_1 is much shorter than reported for high-purity bulk GaAs,[8] but in the presence of free electrons, the relaxation via $A/2(I_+S_- + I_-S_+)$ (Korringa relaxation) reduces T_1.[2,9] Although, on the time scale of our experiments, the memory of the sample is not canceled even if one sweeps the magnetic field to zero, reverses the field direction, and sweeps up again. The nuclear spins remain polarized: $\langle I \rangle \approx$ const, thermal equilibrium is not reached.

From the strength of the Overhauser shift the achieved nuclear-spin polarization can be estimated. Following Eq. (4), B_N in the presence of different nuclei j can be calculated by averaging over all isotopes with respect to their abundance $a^{(j)}$:

$$B_N = \frac{8\pi}{3}\frac{g_0}{g}\mu_N \sum_j a^{(j)} g_N^{(j)} |\psi(r^{(j)})|^2 \langle I^{(j)} \rangle. \quad (5)$$

In the case of bulk GaAs the respective contributions of each isotope are listed in Table I. A maximal nuclear-spin polarization of $\langle I \rangle = -\frac{3}{2}$ for all isotopes yields a maximal Overhauser shift of $B_N \approx 5.3$ T in bulk GaAs.[10] Beyond the value reached in Fig. 2 ($B_N \approx 0.2$ T) we were able to shift the ESR by $B_N \approx 0.43$ T. This shift was limited only by the fact that the sensitivity of ρ_{xx} on ESR is strongly decreased on approaching an even filling factor.[11] Assuming the spin polarization of all isotopes are equal and taking the bulk GaAs value of Table I, we have thus achieved a dynamic nuclear-spin polarization of $\langle I \rangle = -0.12$.

For the determination of the electronic g factors the Overhauser shift is a serious complication, because the measured resonance position is only given by the g factor if $\langle I \rangle = 0$. There are two ways to remove a nuclear-spin polarization: The first is simply to wait for the equilibrium value of $\langle I \rangle$ to be reached. At high magnetic fields the lifetime of a nonequilibrium nuclear-spin polarization is too long for practical purposes. At $B = 0$, however, the hyperfine-interaction-induced flip-flop in the opposite direction opens a relaxation channel for the nuclei, and after a short period of time the nuclear spins are again in equilibrium. The second way to destroy the nuclear-spin polarization involves saturation of the NMR. For this purpose the sample was positioned within a small coil producing ac fields in the MHz range. By performing NMR on one of the particular isotopes (cf. Table I) we were able to destroy the nuclear-spin polarization and to quench the Overhauser shift. Results from such an experiment are shown in Fig. 3. The solid trace shows ρ_{xx} around filling factor $i = 3$ during the first up sweep of the magnetic field ($\langle I \rangle \approx 0$). ESR occurs at $B \approx 5.6$ T. All the other traces are taken while sweeping down, the samples being continuously irradiated not only with microwaves but also with radio frequencies (rf) around $\nu = 40$ MHz. The ESR line is locked to the applied magnetic field (Overhauser shift), resulting, as in Fig. 1, in a resonantly enhanced value of ρ_{xx}. At the magnetic field where the applied rf leads to NMR of, in this case, the ^{75}As isotope, the shifted ESR line is no longer locked to the applied magnetic field ($\dot{B} < 0$) but jumps back to a higher magnetic field as a result of the reduction of $\langle I \rangle$. This is visible in an abrupt change of ρ_{xx} from the resonant to the nonresonant value. Figure 3 shows a series of such measurements at different NMR frequencies. The inset in Fig. 3 gives the magnetic field

TABLE I. A list of the three isotopes in GaAs, with their respective spins I, abundances a, and NMR frequencies ν_{NMR}. The last column gives the contribution of each isotope to the Overhauser shift in bulk GaAs (after Ref. 10).

	I	a (%)	ν_{NMR} at $B = 5.5$ T (MHz)	B_N (T)
^{69}Ga	$\frac{3}{2}$	60.4	56.21	$-0.91\langle I^{69Ga} \rangle$
^{71}Ga	$\frac{3}{2}$	39.6	71.41	$-0.78\langle I^{71Ga} \rangle$
^{75}As	$\frac{3}{2}$	100	40.12	$-1.84\langle I^{75As} \rangle$

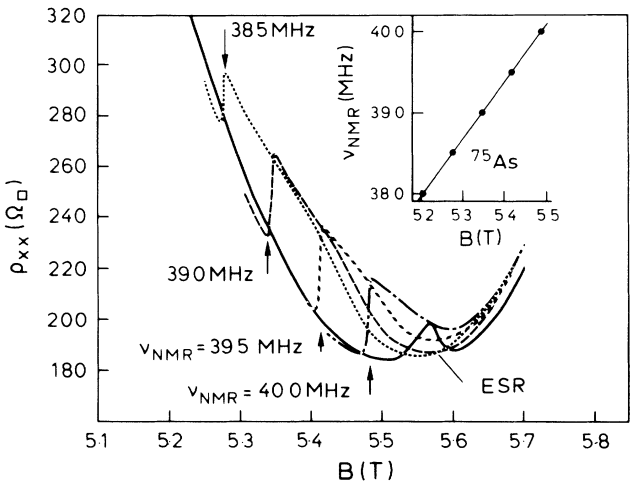

FIG. 3. Magnetoresistivity ρ_{xx} in the vicinity of filling factor $i = 3$ under continuous microwave ($\nu_{ESR} = 23.4$ GHz) and radio-frequency radiation. Solid trace, $\dot{B} > 0$; other traces, $\dot{B} < 0$. The NMR of the ^{75}As nuclei results in a sharp decrease of ρ_{xx} (cf. text). Measurements for different NMR frequencies ν_{NMR} are shown. Inset: magnetic field dependence of ν_{NMR}.

dependence of the sharp decrease in ρ_{xx}, which reflects the well-known magnetic field dependence of the ^{75}As nuclear-spin splitting: $\nu_{NMR}(B) = (7.295\ \text{MHz/T})B$. The rf coil designed for the ^{75}As frequencies happened to operate also at frequencies around 70 MHz. This also permitted the ^{71}Ga NMR to be observed (not shown).

In summary, we have for the first time seen a direct interaction between two-dimensional electrons and nuclei. From the Overhauser shift we can conclude an extraordinarily high nuclear-spin polarization, which allows the NMR study of all polarized nuclei, i.e., those in the direct vicinity of the two-dimensional electron gas. By applying radio frequencies to GaAs-$Al_xGa_{1-x}As$ heterostructures we have been able to detect nuclear magnetic resonance in the magnetoresistivity.

We would like to thank H. Obloh for valuable discussions, J. Behring and G. Wilk for expert technical help, and A. Plaut for a careful reading of the manuscript. This work has been supported by the Stiftung Volkswagenwerk and the Bundesministerium für Forschung und Technologie.

[1]M. Dobers, K. v. Klitzing, and G. Weimann, Phys. Rev. B **38**, 5453 (1988).

[2]A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961), p. 191ff.

[3]G. E. Pake and T. L. Estle, *The Physical Principles of Electron Paramagnetic Resonance* (Benjamin, Reading, MA, 1973), 2nd ed., p. 109.

[4]A. W. Overhauser, Phys. Rev. **92**, 411 (1953).

[5]M. Guéron and Ch. Ryter, Phys. Rev. Lett. **3**, 338 (1959).

[6]For a review see D. Paget and V. L. Berkovits, in *Optical Orientation,* edited by F. Meier and B. P. Zakharchenya, Modern Problems in Condensed Matter Sciences Vol. 8 (Elsevier, New York, 1984), p. 381.

[7]G. Denninger, W. Stöcklein, E. Dormann, and M. Schwoerer, Chem. Phys. Lett. **107**, 222 (1984).

[8]J. A. McNeil and W. G. Clark, Phys. Rev. B **13**, 4705 (1976).

[9]I. D. Vagner, T. Maniv, and E. Ehrenfreund, Solid State Commun. **44**, 635 (1982).

[10]D. Paget, G. Lampel, B. Sapoval, and V. S. Safarov, Phys. Rev. B **15**, 5780 (1977).

[11]D. Stein, G. Ebert, K. v. Klitzing, and G. Weimann, Surf. Sci. **142**, 406 (1984).