Pergamon

Solid State Communications, Vol. 102, No. 10, pp. 715–720, 1997

0038-1098/97 $17.00+.00

PII: S0038-1098(97)00101-4

NUCLEAR SPIN RELAXATION IN AlGaAs/GaAs HETEROSTRUCTURES OBSERVED VIA OPTICALLY DETECTED MAGNETIC RESONANCE (ODMR) EXPERIMENTS

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(*Received* 28 *January* 1997; *accepted* 10 *March* 1997 *by M. Cardona*)

The resonant field at which an electron spin resonance (ESR) occurs may be shifted by an effective magnetic field, which is due to spin polarized nuclei. This shift, known as Overhausershift, is caused by the field of all polarized nuclei without respect to their isotopic number. Irradiation of the nuclear magnetic resonance (NMR) frequency of some of the isotopes during the ESR experiment allows to measure the nuclear field of only one kind. The time dependent evolution of the Overhausershift is according to the nuclear spin relaxation time. In undoped semiconductors e.g. in AlGaAs/GaAs heterostructures, typical time constants are determined to be between 10 minutes and one hour; irradiation with visible light which changes the number of free carriers affects the relaxation process. We further show that the ratio of the relaxation times of the two Ga-isotopes ^{69}Ga and ^{71}Ga behaves as a quadrupolar relaxation rather than relaxation via Fermi contact interaction with free carriers. This result is supported by optically detected NMR measurements, which exhibit a quadrupolar splitting of the NMR resonance line. ©1997 Elsevier Science Ltd

1. INTRODUCTION

The optical detection of ESR in AlGaAs/GaAs heterostructures is done by measuring the degree of polarization of the photoluminescence. By excitation of the sample with a circularly polarized laser beam at an energy slightly above the energy gap of GaAs, the excited electrons in the conduction band will be partially spin polarized as well due to different excitation probabilities for heavy and light hole states [1]. The maximum electronic spin polarization is up to 100% if the degeneracy of these valence band states is lifted and the laser energy is small enough to excite only electrons from the upper heavy hole valence band. In our experiments the laser had a higher energy, so the initial polarization was only 50%. This polarization decreases during the lifetime of the electrons and the measured degree of photoluminescence polarization depends on the ratio of electron spin relaxation time to electron lifetime in the conduction band [2]. Electron spin relaxation has many reasons [3] one of them is the hyperfine coupling of the electron spin to the nuclear spin. The hyperfine interaction $\hat{H}_{SI}$ consists of two terms, the dipole interaction $\hat{H}_{dip}$, which is of less importance in cubic crystals like GaAs, and the Fermi contact interaction $\hat{H}_F$:

$$\hat{H}_{SI} = \hat{H}_F + \hat{H}_{dip} \tag{1}$$

where

$$\hat{H}_F = \hat{S}\tilde{A}\hat{I} \tag{2}$$

This part arises due to a non vanishing expectation probability of electrons at the nuclear site $|\psi(r = 0)|^2$. $\tilde{A}$ is the tensor of the hyperfine interaction, and may be written as a scalar for cubic crystals. $\hat{S}$ and $\hat{I}$ are the operators for electron and nuclear spin, respectively. Applying a magnetic field in the direction of z one gets

$$\hat{S}A\hat{I} = A(S_xI_x + S_yI_y + S_zI_z) \tag{3}$$

which may be written as

$$\hat{S}A\hat{I} = \frac{1}{2}A(\hat{I}_+\hat{S}_- + \hat{I}_-\hat{S}_+) + AI_zS_z \qquad (4)$$

Here the terms $\hat{I}_\pm\hat{S}_\mp$ describe flip-flop processes, where the z-components of the electron- and nuclear-spin get raised and diminished respectively and vice versa.

Since the spin Hamilton operator of the electrons $\hat{H}_S$ is:

$$\hat{H}_S = g^*\,\mu_B\,\hat{S}_z\,B_0 \qquad (5)$$

the total Hamiltonian describing the electronic energy $\hat{H}_S$ as well as the interaction $\hat{H}_{SI}$ between nuclei and electrons results in

$$H_e = g^*\mu_B B_0\hat{S}_z + A\langle I_z\rangle S_z = g^*\mu_B B_0(1+D)\hat{S}_z \qquad (6)$$

where $\langle I_z\rangle$ is the mean nuclear polarization and D is called Overhausershift

$$D := \frac{A\langle I_z\rangle}{g^*\mu_B B_0} \qquad (7)$$

If the electrons are pumped with circularly polarized light of one sense of revolution the nuclei will be polarized dynamically due to the above mentioned flip–flop processes. Thus the nuclear spin polarization can reach values far above thermal equilibrium due to this dynamic polarization. Since g^* is small in GaAs (−0.44) the Overhausershift D can obtain considerable values which were measured to determine the time dependence of the nuclear polarization.

In an optical experiment the ESR is observed as a resonant decrease of electron spin polarization in the conduction band via the photoluminescence polarization.

The spin relaxation time of nuclei in metals and semiconductors at low temperatures is usually understood as a consequence of the hyperfine interaction. $T_{1,HFI}$ has typical values between 0.1 and 10^4 s [4]. Other relaxation mechanisms $T_{1,o}$ add to the whole relaxation rate as following

$$\frac{1}{T_{1n}} = \frac{1}{T_{1,HFI}} + \frac{1}{T_{1,o}} \qquad (8)$$

This paper presents experimental data, which suggest that quadrupolar relaxation has a substantial influence. Quadrupolar interaction arises due to an electric field gradient, which has internal reasons and may only be changed slightly externally. This is in contrast to magnetic dipole moments which interact with magnetic fields, and thus may be changed by applying an external magnetic field [5]. In the point group T_d (GaAs) the nuclear spin states are $2I + 1$–fold degenerated [6], except there are electrical field gradients existing, which is the case e.g. in the vicinity of lattice defects. Further, for the observation of quadrupole effects the nucleus must posses a nuclear spin greater than 1/2, which is fulfilled for all nuclear species in GaAs (^{69}Ga, ^{71}Ga, ^{75}As) having $I = 3/2$. Their quadrupolar constants Q, taken from [7] show significant values. Since GaAs belongs to T_d quadrupolar effects where not expected for years [8]. But in compositions containing e.g. aluminum, or at the interfaces in heterostructures where the cubic symmetry is broken if Ga–atoms are replaced by Al, quadrupolar effects may be expected without doubt. Quadrupolar effects can result in splittings of NMR–lines and/or change of nuclear–spin–lattice relaxation times.

The energy splitting of a NMR line due to quadrupole interaction can be written as

$$\nu_Q = \frac{3eQV_{zz}}{2I(2I-1)h} \qquad (9)$$

$$\Delta E_Q(m) = \frac{h\nu_Q}{2}(m^2 - \frac{a}{3}) \qquad (10)$$

with

$$a = I(I+1)$$

[8,9], where V_{zz} is the electric field gradient parallel to the external magnetic field, Q the nuclear quadrupole moment and m the magnetic quantum number.

The spin–lattice relaxation time T_1 in the case of quadrupolar relaxation strongly depends on whether being above or below the Debye temperature θ [10] of the investigated material. Theoretical analysis results in [5]:

$$\frac{1}{T_1} \sim T^2(a - \frac{b}{T^2}) \quad T \geq \frac{1}{2}\theta \qquad (a, b = \text{const.})$$

$$\frac{1}{T_1} \sim T^7 \qquad T \leq 0.02\,\theta$$

Experimental results [11] show other dependencies ($\sim T^{9.5}$). Since a precise calculation of relaxation times is not possible a factor γ is introduced to take into account the difference between the applied point–charge model and the reality [12, 13]. In γ the Sternheimer anti–shielding factors are included, which cause an amplification of the electric field gradient. The values of γ are between 10 and 300. The relaxation time T_1 is proportional to γ^{-2}.

A detailed derivation of quadrupole spin lattice relaxation for high temperatures shows a Q^2–dependency [8].

$$\frac{1}{T_1} \sim \frac{9}{8\pi^3}\frac{1}{\hbar^3}\left[\frac{e\gamma Q}{R^3}\right]^2\left[\frac{k_BT}{mv^5}\right]\omega_m^5 \qquad (11)$$

Q is the electric quadrupole moment, the other parameters are of no further interest. The proportionality for low temperatures ($1/T_1 \sim T^7$) is to our opinion not applicable to the data presented here and will

be discussed later in detail as well as the fact that a Q^2 dependence is observed regardless of temperatures well below θ in our experiments.

2. EXPERIMENTAL RESULTS

The experiments were done at low temperatures using a bath cryostat with optical access to the sample which is immersed in liquid helium pumped down to 1.8 K. The magnetic field was applied with a Bruker BE–10 conventional magnet. The samples were excited with a laser diode at a wavelength of 757 nm, the exciting light could be either modulated left/right–circular or held at constant circular polarization. The photoluminescence is analyzed circular polarization sensitive, through a 64 cm monochromator and detected with a GaAs–photomultiplier. The photon counter is gated with the modulation signal of the σ^+/σ^- modulator ($\approx$1 kHz). Under modulated excitation it is possible to measure photoluminescence polarization without creating a dynamic nuclear polarization due to the high modulation frequency with respect to the slow nuclear spin relaxation rate.

For relaxation experiments the sample was first irradiated with a constant circularly polarized light for dynamic nuclear polarization and then the modulator was switched on to observe the relaxation of the nuclear spin polarization to thermal equilibrium.

The dielectric ESR resonator is homemade and specially designed for ESR/NMR double resonance experiments. It allows optical access to the sample. A small loop in the cavity enables irradiation of radiofrequency (rf) to saturate nuclei selectively by NMR. The measured quality factor is only about 1400, but due to the small volume the $\frac{B_1^{\text{rot}}}{\sqrt{P}}$ is about $0.2\frac{\text{mT}}{\sqrt{\text{W}}}$. The resonant frequency is about 7.3 GHz and a maximum power of 1 W could be applied. For selective measurement of relaxation times of one kind of nuclei the other species were depolarized by rf pulses with their NMR frequency during the pass of the ESR resonance. So no nuclear polarization effects of others than the desired isotope were detected.

Three different samples were investigated. A p–channel AlGaAs/GaAs heterostructure, which has a two–dimensional hole gas (2DHG) at the interface, a multiquantumwell (MQW) sample, AlGaAs/GaAs with 10 quantum wells of 10 nm well width and a quantum dot sample, which is primarily a modulation doped AlGaAs/GaAs heterostructure with a two–dimensional electron gas (2DEG), but due to an etched grid pattern on the surface, the 2DEG potential is modulated such, that a quasi quantum dot structure is achieved.

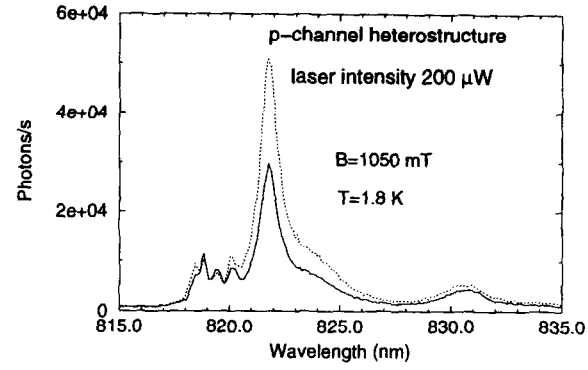

Fig. 1. Photoluminescence of the p-channel heterostructure. The two traces represent the intensity of left and right hand circularly polarized light. Excitation was circularly polarized; laser wavelength 757 nm.

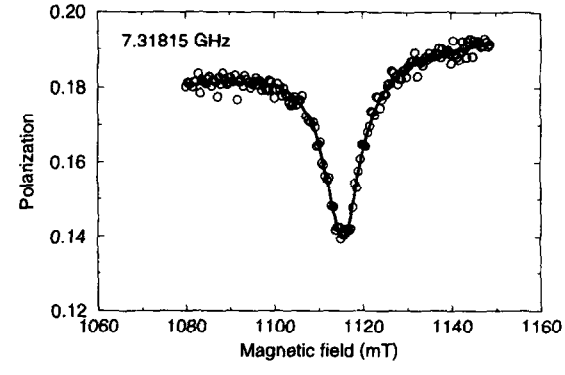

Fig. 2. Typical ODESR curve of p–channel heterostructure with a Lorentzian fitted to the experimental data. Microwave frequency: 7.31815 GHz.

Figure 1 shows exemplary for the 3 samples a typical photoluminescence spectrum of the p–channel heterostructure. The two traces represent the different circular polarizations (σ^+, σ^-). The peak at 822 nm is identified as the so called H-band [14] and shows a significant polarization, which is detected in the ODNMR and ODESR experiments.

Figure 2 shows a typical ODESR measurement at the p–channel heterostructure with a Lorentzian fitted to the experimental data. Thus the determination of the resonance position is possible with great accuracy.

Figure 3 shows a relaxation curve for the ^{75}As–nuclei in the p–channel heterostructure. The solid line is a mono exponential fit to the experimental data. At $t = 0$ the modulator was activated and the relaxation to the equilibrium position started. For the particular experimental conditions the fit gives a relaxation time of 12.9 min.

It is worth to point out that the mono exponen-

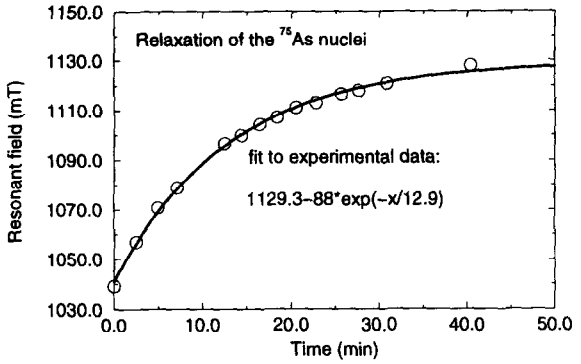

Fig. 3. Observation of the nuclear spin relaxation by detecting the Overhausershift of the ESR. The mono exponential fit shows a relaxation time T_1^{75As} of 12.9 min for the ^{75}As nuclei.

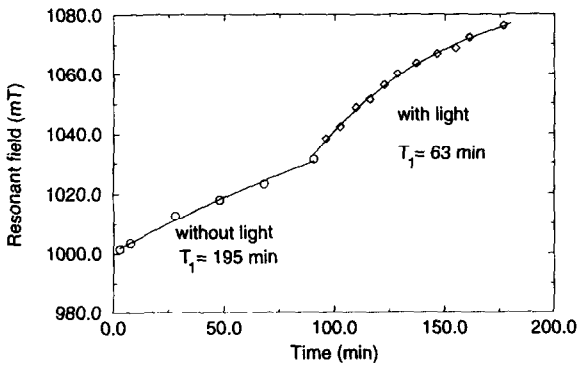

Fig. 4. Comparison of relaxation times with and without light irradiation. From t=0 to t=80 min the sample was only irradiated for times as short as possible for the measurement, after t=80 min there was a constant irradiation with 0.5 mW.

tial relaxation is observed if only one nuclear species is measured. The measured relaxation time may be a combination of different mechanisms, e.g. quadrupolar relaxation effects and Fermi–contact interaction may be mixed. This important fact can be seen in Fig. 4, where the sample was irradiated with different laser powers. In the region from 0 to 80 min the sample was illuminated only as shortly as possible to make the optical detection measurement, but most of the time relaxation took place without light. The second part, after 80 min was measured with constant illumination with a power of 0.5 mW. The experiment exhibits a factor 3 slower nuclear spin relaxation time without light. The effect is readily explained with relaxation via the Fermi–contact interaction, which needs photo generated electrons in the conduction band for its mechanism. If no light is irradiated, no electrons are excited and the mechanism fails, causing a smaller relaxation rate.

Table 1. Results of the relaxation time measurements of the three different samples. (All times in minutes).

	^{75}As	^{69}Ga	^{71}Ga
QD-sample	20.8	6.8	20.1
p-channel heterostructure	11.7	9.8	25.1
MQW-sample	50.8	23.1	42.1

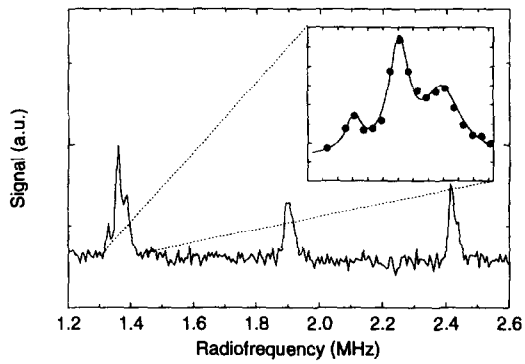

Fig. 5. Quadrupole splitting of the ODNMR ^{75}As–line, the insert shows a fit of 3 Lorentzians to the splitted line. The other resonance lines of ^{69}Ga at 1.9 MHz and ^{71}Ga at 2.4 MHz do not exhibit a significant splitting.

Due to this reason values of relaxation times in Tab. 1 are exact for comparison of the relative relaxation times of different nuclear species, since the three times of each sample were taken under same conditions (laser power and spot size, temperature), but only describe times under special conditions and at a certain position on the sample surface.

ODNMR measurements on the p–channel heterostructure revealed a splitting of the ^{75}As–resonance line in up to 3 components as expected from the quadrupole splitting (see Equ. [10]). Figure 5 shows the entire NMR spectrum with the splitted 3–fold ^{75}As line and the two Ga isotope resonance lines (^{69}Ga, ^{71}Ga).

3. DISCUSSION

The nuclear spin splitting of ^{75}As is fitted with 3 Lorentzians, which show a frequency split ν_Q = 29 kHz. Since the experiment only measures the electric field gradient parallel to the external field B_0 only V_{zz} is obtained. With Equ. [10] one gets

$$V_{zz} = 8.3 \cdot 10^{18} \frac{\mathrm{V}}{\mathrm{m}^2}$$

This result is in good agreement with the one of Marohn et al. [15] where $6.76 \cdot 10^{18} \frac{\mathrm{V}}{\mathrm{m}^2}$ were found in

Table 2. Comparison of the relaxation time ratios $T_1^{69}Ga/T_1^{71}Ga$

	$T_1^{69}Ga/T_1^{71}Ga$
QD–sample	0.34
p–channel heterostructure	0.39
MQW–sample	0.55

a comparable sample with another experimental technique. The quadrupole moment of the ^{75}As nucleus is $Q = 0.29 \cdot 10^{-28}$ m^2 [7]. It is not surprising that the other resonance lines of the Ga–nuclei do not show a pronounced splitting, since their quadrupole moments are smaller and so it is not resolved. This result shows that the tetrahedral symmetry at the nuclear site is broken, which might be due to local effects (defects) e.g. from lattice mismatch in the vicinity of the AlGaAs/GaAs interface.

Discussing the relaxation times in semiconductors a Fermi–contact exchange interaction is readily assumed. Since here $1/T_1 \sim \gamma_n^2$, γ_n being the gyromagnetic ratio of the nucleus of kind n, we can calculate the expected ratio of the nuclear relaxation times. Because all other parameters like $|\psi(0)|^2$ should be equal for isotopes of one species as ^{69}Ga and ^{71}Ga we get:

$$\frac{T_1^{69}Ga}{T_1^{71}Ga} = \left(\frac{\gamma_n^{71}Ga}{\gamma_n^{69}Ga}\right)^2 \approx 1.6$$

This means that the $T_1^{69}Ga$ should be longer than $T_1^{71}Ga$ by a factor 1.6. Experimental data show that the $T_1^{69}Ga$ is shorter!

Table 2 lists the experimentally found ratios for the 3 investigated samples.

Assuming a quadrupolar relaxation Equ. [11] yields for the relaxation rate:

$$\frac{1}{T_1} \sim Q^2 \qquad (12)$$

which means

$$\frac{T_1^{69}Ga}{T_1^{71}Ga} = \left(\frac{Q^{71}Ga}{Q^{69}Ga}\right)^2 = \left(\frac{0.106}{0.168}\right)^2 \approx 0.4 \quad .$$

This ratio is much closer to the experimental findings. Deviations of the observed values from this theoretical expectations may be due to other relaxation mechanisms or due to small temperature shifts during the time of about 1 hour necessary between relaxation measurements for different nuclei. If one accepts the T^7 or $T^{9.5}$ dependencies stated in other publications, even smallest deviations could have significant influence on the relaxation time.

These high exponents originally let us expect a relatively small influence of quadrupolar relaxation at low temperatures which is obviously not the case. It seems that effects due to defects cause bigger influences than considered up to now.

4. CONCLUSION

The paper shows measurements of nuclear spin lattice relaxation times on different AlGaAs/GaAs samples. The ratio of the relaxation times of the two Ga isotopes gives strong evidence to a dominance of quadrupolar relaxation mechanisms. This statement is supported by an ODNMR measurement which reveals a quadrupole splitting of the NMR resonance line which is due to a significant electrical field gradient.

Since the ODESR is measured with a laser, focused to about 200 μm diameter it is possible to scan the sample surface and find sites with high defect densities.

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