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Nuclear spin relaxation mediated by Fermi-edge electrons in *n*-type GaAs

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A method based on the optical orientation technique was developed to measure the nuclear-spin lattice relaxation time T_1 in semiconductors. It was applied to bulk n-type GaAs, where T_1 was measured after switching off the optical excitation in magnetic fields from 400 to 1200 G at low (< 30 K) temperatures. The spin-lattice relaxation of nuclei in the studied sample with $n_D = 9 \cdot 10^{16}$ cm^{-3} was found to be determined by hyperfine scattering of itinerant electrons (Korringa mechanism) which predicts invariability of T_1 with the change of magnetic field and linear dependence of the relaxation rate on temperature. This result extends the experimentally verified applicability of the Korringa relaxation law in degenerate semiconductors, previously studied in strong magnetic fields (several Tesla), to the moderate field range.

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1. Introduction. First experiment on optical orientation of electron and nuclear spins in semiconductors was conducted more than 40 years ago [1]. Since then, spin dynamics in electron-nuclear spin systems was a subject of many theoretical and experimental works. Following the estimation by Dyakonov and Perel of the rate of electron-nuclear spin transfer mediated by localized and delocalized electrons it was believed for a long time that the latter cannot provide effective channels neither for dynamic polarization of nuclear spins nor for their relaxation [2]. However, in heavily-doped n-type semiconductors the density of electron states at the Fermi level can be high enough to make the Korringa spin relaxation mechanism competitive to other nuclear spin relaxation channels. The Korringa mechanism, i.e. spin relaxation of lattice nuclei via scattering of itinerant electrons between states in the vicinity of the Fermi surface was experimentally observed in many metals [3]. Its fingerprint is linear dependence of the relaxation rate on temperature, owing to the fact that the number of electrons with unpaired spins is proportional to the kinetic temperature of the electron gas.

Korringa mechanism was documented at high magnetic fields (> 1 T) for several heavily doped semiconductors, including Si [4], Ge [5] and GaAs and InP [6, 7]. On the other hand, in their recent paper, Kolbl et al. reported a breakdown of Korringa law of nuclear spin relaxation at temperatures below 1 K and weak magnetic field (15 G) [8]. Also, a contribution of localized electron states into nuclear spin relaxation was found in just-metallic ($n_D = 2 \cdot 10^{16}$ cm^{-3}) GaAs [9], also at low magnetic fields (< 200 G). In both these works, specially designed structures were used to detect the nuclear magnetization either by the spin-injection technique [8], or by Faraday rotation in a high finesse microcavity [9].

Main goals of our work were to investigate the nuclear spin dynamic in metallic phase of bulk n-type GaAs in the intermediate range of magnetic fields (between 10 mT and 1 T), to find out if the Korringa relaxation remains the dominating mechanism in this range, and to develop a method based on optical orientation technique, which would be applicable for studying various types of semiconductor structures and could be extended to a wider range of magnetic fields and temperatures. The choice of GaAs for this study is justified by its prospective spintronic applications considering there is an idea of using nuclear spin for information storage and processing [10, 11]. Also, since the physics of spin systems in GaAs is very well studied, once an understanding of the spin-relaxation processes is reached for this semiconductor it can be easily extended to other semiconductors.

All the measurements were done using optical orientation method at various low magnetic fields (400, 800, and 1200 G) and low temperatures (from 2 up to 30 K). In our experimental protocol processes of optical cooling of nuclear spin system and measurement of its spin temperature were divided in time similar to the procedure

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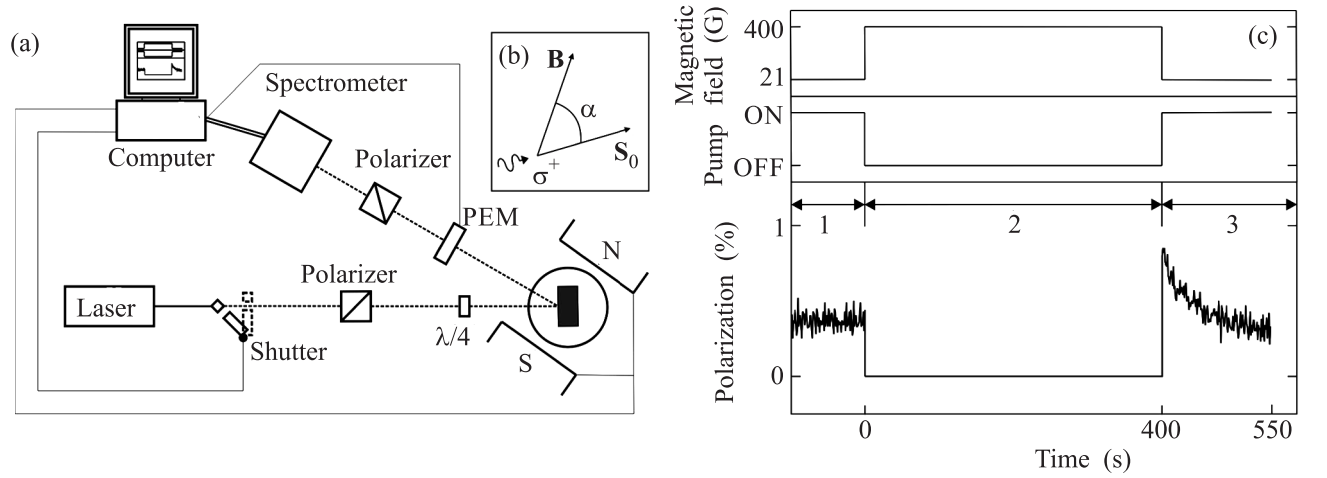

Fig. 1. (a) – Schematic diagram of experimental setup. Armoured electromagnet was used to apply external magnetic field. Both mechanical shutter and power source for the magnetic field were controlled by the computer which allowed that shutter switching on/off could be followed by change of magnetic field. (b) – Schematic representation of oblique geometry where magnetic field is oriented at a small angle with incident beam. (c) – Experimental procedure time evolution: *1* – dynamic polarization of nuclei with circularly polarized light, *2* – spin-lattice relaxation of nuclei during dark period when different magnetic fields were applied, *3* – detection of nuclear polarization via depolarization of luminescence in measuring magnetic field

first realized by Kalevich et al. [12]. During the pumping stage, circularly-polarized light with photon energy just above the bandgap of GaAs was shone on the sample, so that nuclear spins were being dynamically polarized via interaction with optically oriented electrons. During the "dark" stage the pumping light was switched off, and the nuclear spins were let to relax in the magnetic field of interest during a variable time interval. The nuclear polarization to remain after the "dark" interval was detected at the beginning of the next pumping stage using the Hanle effect modified by the Overhauser field of spin-polarizad nuclei [13]. To this end, the magnetic field was applied at an oblique angle to the excitation laser beam. During the pumping stage, the field strength was set to the value where the electron mean spin was most sensitive to variations of the nuclear field.

2. Experimental technique. We used a bulk n-type GaAs sample with doping concentration of $n_D = 9 \cdot 10^{16}\,\mathrm{cm}^{-3}$ (metallic phase). Sample was grown by the molecular-beam epitaxy (MBE) as a $2\,\mu$m thick layer of GaAs between AlGaAs barriers. All measurements were done at cryogenic temperatures using two different types of cryostats: a cryostat cooled by liquid helium that operates at 2 and 4.2 K and a two stage compressor-cooled cryostat for temperatures from 10 up to 30 K. Injection of spin polarized electrons has been achieved by pumping the sample with light from a tunable Ti-sapphire laser. Circularly polarized light with the photon energy just above E_g of GaAs excited spin-oriented electrons in the sample. Photoluminescence (PL) was collected in the reflection geometry. The combination of a photoelastic modulator (PEM) and a linear polarizer passed alternatively the right-hand and left-hand polarized PL through a double-grating spectrometer to an avalanche photodiode, connected with a two-channel photon counter synchronized with the PEM. In this manner we could register quantity of N_+ (right-hand) and N_- (left-hand) oriented photons and derive degree of circular polarization $\rho = (N_+ - N_-)/(N_+ + N_-)$.

In our experiment we have applied oblique external magnetic field in the near-Voigt geometry, i.e. at a small (< 10 deg) angle to the sample plane.

On the time scale, our experimental protocol consisted of a sequence of "bright" and "dark" intervals. During the "bright" stage (Fig. 1c, *1*), cw circular polarized light (excitation energy 1.55 eV) excited spin-polarized electron-hole pairs, and this way provided injection of non-equilibrium spin in the ensemble of resident electrons [13]. When electrons have non-equilibrium spin polarization, hyperfine interaction results in dynamic polarization of nuclei, as a measure of the electron spin projection onto the direction of the oblique field. After initial polarization of the sample, pumping was switched off, which was followed by instant change of magnetic filed (Fig. 1c, *2*). Since there was no injection of spin polarized electrons into the system, electron spin polarization very quickly (nanoseconds) returned to its equilibrium state, and nuclear spin polarization started to relax due to interaction of nuclei with unpolarized elec-

tron bath. By the end of the dark stage it decreased by the factor of e^{-t/T_1}, where t is the duration of the dark interval. During the second bright stage (Fig. 1c, *3*) measuring field was restored and we detected circular polarization of the PL, determined by the dynamic equilibrium of pumping, spin relaxation, and Larmor precession of electron spins around the effective field, equal to the sum of external field and nuclear magnetic field [13]. As the nuclear field increased in the course of dynamic polarization of nuclei, PL polarization changing in time could be observed. Since the projection of the injected electron spin on the external field was small, the Overhauser field did not exceed several Gauss, and it was possible to choose a point on the Hanle curve where the dependence of the PL polarization on the Overhauser field was to a good precision linear. In this regime we observe the polarization signal exponentially approaching some equilibrium value, with the time decrement about 50 s. We used 150 s long bright intervals to allow the nuclear polarization to come to saturation. The starting point of this time evolution depended on the pre-history. If the sample had been kept in darkness for a long time (in our case, of the order of 20 min), it corresponded to zero Overhauser field, i.e. the PL polarization was determined by the Hanle effect in the external field only. If, however, the duration of the preceding dark interval was comparable to the nuclear spin-lattice relaxation time T_1, the Overhauser field at the beginning of bright interval was not zero.

In the linear regime, when the difference in nuclear fields, ΔB_N that develops during the dark time, is small, the difference between polarization degrees at the moments of light switch off, ρ^{off} and switch on, ρ^{on} is proportional to $\Delta\rho = \rho^{\text{off}} - \rho^{\text{on}} \sim \Delta B_N \sim e^{-t/T_1}$. Fitting polarization of luminescence as a function of time with an exponentially decaying function, we can determine the polarization degree at the point when the pump was switched back on (the highest point on the exponential curve), ρ_t.

In order to relate the observed changes in the PL polarization with the time dependence of the Overhauser field, we measured the electronic Hanle curve. This was done by putting the PEM, set to $\lambda/4$ retardance, in the excitation channel. Modulation of exciting beam from σ^+ to σ^- polarization at 50 kHz allowed us to free ourselves from the influence of nuclear polarization which is strongly suppressed, since it cannot follow this fast change of the mean electron spin. In our sample, depolarization of electron spin in external magnetic field was well described by the Lorentz curve:

$$s_z(B) = s_z(0)\frac{1}{1+(\mu_{\text{B}}gBT_s/\hbar)^2}, \qquad (1)$$

where s_z is the projection of mean electron spin on the direction of excitation, μ_{B} is the Bohr magneton, g is the electron g-factor and $B = B_{\text{ext}} + B_N$ is the effective magnetic field. Considering that Hanle curve half width $B_{1/2}$ is equal to the $(\hbar/\mu_{\text{B}}g)T_s^{-1}$ and that in III–V semiconductors $\rho = s_z$ we can rewrite Eq. (1) as $\rho_t = \rho_0/\{1 + [(B_{\text{ext}} + B_N)/B_{1/2}]^2\}$, where ρ_0 is the degree of circular polarization in the absence of the external magnetic field. From here we can obtain equation for nuclear magnetic field:

$$B_N = B_{1/2}\sqrt{\frac{\rho_0 - \rho_t}{\rho_t}} - B_{\text{ext}}. \qquad (2)$$

Using this equation we can calculate the change of nuclear field as a function of darkness time (time when there is no optical pumping). It declines exponentially as the dark interval becomes larger and it is proportional to e^{-t/T_1}. Longitudinal relaxation time, T_1 can be obtained by fitting this change with exponentially decaying function.

In order to determine magnetic field dependence of T_1, laser cut off was followed by change in external magnetic field, so that during dark intervals various fields (400, 800, and 1200 G) were engaged. A measurement of T_1 magnetic field and temperature dependence hence consists of series of measurements with varying B and T but keeping all the others variables the same. In order to increase accuracy of experiment each measurement consisted from several passes and all the results were averaged.

3. Results. The photoluminescence intensity spectrum (Fig. 2) consists of two peaks, one corresponding to

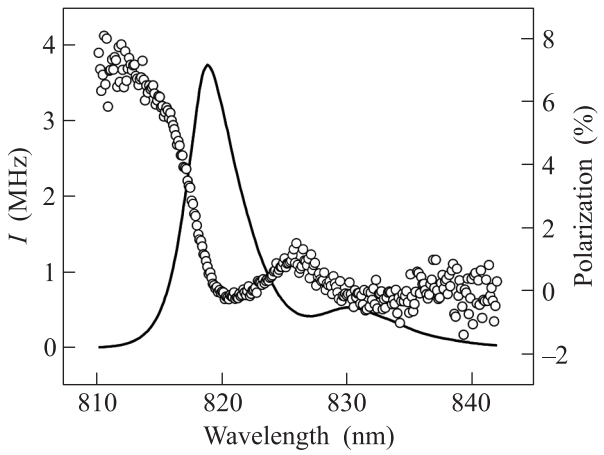

Fig. 2. Photoluminescence spectra of n-GaAs with $n_D = 9 \cdot 10^{16}\,\text{cm}^{-3}$: intensity (solid line) and circular polarization of photoluminescence (circles). Nuclear spin relaxation measurments were done at detection setings at 816 nm

the recombination of excitonic complex (819.1 nm) and second one being connected with the optical transition to the shallow acceptor level (830.6 nm) [14].

Change of polarization in transverse magnetic field obtained by exciting the sample with modulated light is presented in Fig. 3 (squares). In this case nuclear polarization is strongly suppressed, thus Hanle curve is symmetric with respect to zero field and well described by Lorentz function. Depolarization of luminescence in Fig. 3 (circles) was measured at constant circular polarization of the exciting light. In this case the Hanle curve has the typical asymmetric shape owed to effect of the Overhauser field [13]. For our experiments we chose the steeper slope of the Hanle curve, where the Overhauser field adds up to the external magnetic field.

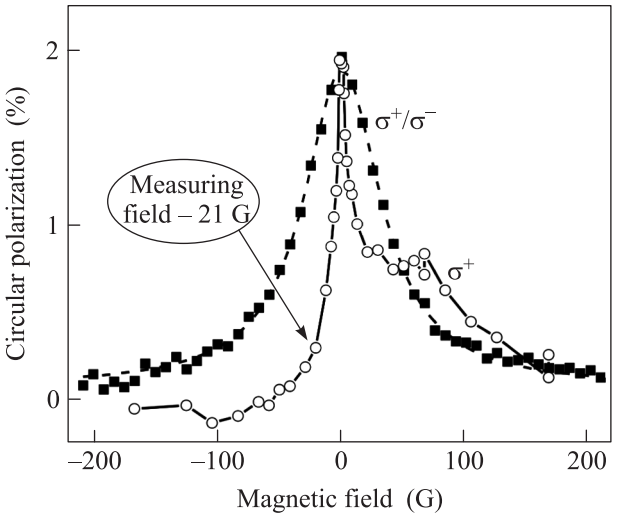

Fig. 3. Magnetic depolarization of luminescence at constant polarization of light (circles). Depolarization of luminescence (squares) fitted with Lorentzian at σ^+/σ^- alternation of exciting light at 50 kHz. Measuring temperature was 10 K

The magnetic field dependence of T_1 was obtained by applying different magnetic fields (400, 800, and 1200 G) during the period when pump was switched off (Fig. 4a). It can be seen (Fig. 4a) that there is no significant variation in relaxation time within this range of magnetic fields. Moreover, our data are very close to those measured by Lu et al. [6] in a GaAs sample with similar doping ($n_D = 7 \cdot 10^{16}\,\mathrm{cm}^{-3}$) at much stronger field (15 kG). This fact agrees well with the suggestion that the nuclear spin-lattice relaxation is determined by Korringa mechanism. The Korringa mechanism can be understood as an effect of the fluctuating filed produced by conduction electrons at the nucleus. Because of rapid movements of electrons at the Fermi surface, the correlation time of this random field is very short as compared to Larmor or cyclotron periods of electrons in this range of magnetic fields. For this reason, the Korringa mechanism is field independent in a wide range of magnetic fields [3]. Its temperature dependence follows from the fact that, microscopically, it is a result of flip-flop transitions of nuclear spins and spins of itinerant electrons near the Fermi surface. Energy, $\hbar(\omega_e - \omega_N)$, needed for this transition to occur is provided by changing the electron kinetic energy (here $\omega_e = -\gamma_e B_0$ and $\omega_N = -\gamma_N B_0$ are the electron and nuclear Larmor frequencies). Since the average kinetic energy of electrons is much larger compared to the thermal energy kT and is of the same order of magnitude as the Fermy energy E_F ($E_\mathrm{F} = \frac{\hbar^2}{2m_e^*}(3\pi^2 N_D)^{2/3} = 0.01\,\mathrm{eV}$) many conduction electrons cannot take or give up the small energy $\hbar(\omega_e - \omega_N)$ because of Pauli principle. This means that only electrons within kT of the Fermi surface can participate in the nuclear relaxation process. In the free-electron approximation nuclear relaxation rate is given by:

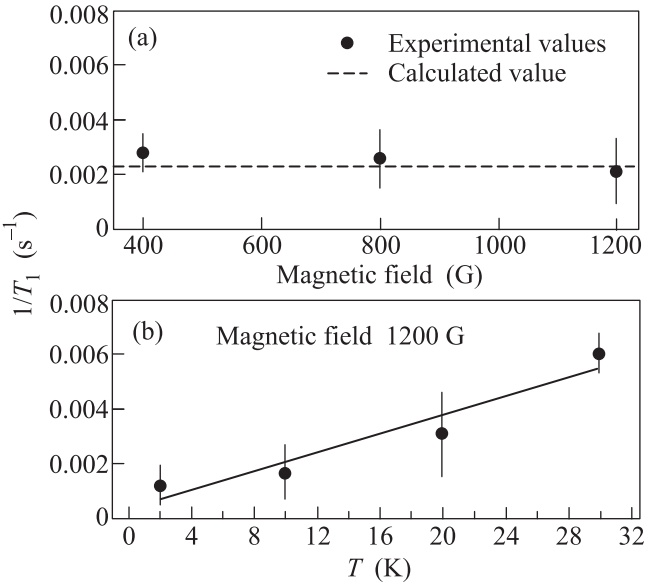

Fig. 4. (a) – Magnetic field dependence of reciprocal nuclear spin lifetime recorded at 10 K. Dashed line corresponds to $1/T_1$ calculated value. (b) – Temperature dependence of reciprocal nuclear spin lifetime recorded at 1200 G external magnetic field

$$\frac{1}{T_1} = \frac{\pi}{\hbar} A^2 v_0^2 \rho^2(E_\mathrm{F}) kT, \qquad (3)$$

where $A = \frac{8\pi}{3}\gamma_e\gamma_N\hbar^2$ is the hyperfine coupling constant (a value of 44 μeV has been used which lies in between a hyperfine coupling constants for Ga (42 μeV) and As (46 μeV)), v_0^2 is the primitive cell volume for fcc, and $\rho^2(E_\mathrm{F})$ is the density of states at Fermy level. Using Eq. (3) we calculated that the spin-lattice relaxation

time for our sample (472 s at 10 K) and the agreement with all the measured points is more than satisfying. Since Korringa mechanism predicts linear dependence of $1/T^{-1}$ on temperature we also did a measurement where we kept external magnetic field constant (1200 G) and change the temperatures inside the cryostat (from 2 to 30 K). Indeed, the temperature dependence of $1/T_1$ at 1200 G (Fig. 4b) can be approximated, within our experimental precision, by a linear function with the slope predicted by the Korringa law and the intercept close to zero.

To conclude, nuclear-spin lattice relaxation time was measured in degenerate bulk n-type GaAs at low temperature and magnetic field range from 400 up to 1200 G. It has been found that Korringa relaxation process remains dominant mechanism for relaxation of nuclear spins at moderate range of magnetic fields, a fact already observed in many heavily-doped semiconductors at high magnetic fields (< 1 T). Also, a method suitable for studying nuclear spin dynamics, its temperature and magnetic field dependence in various semiconductors was developed from optical orientation modus.

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