Manifestation of dipole-dipole and quadrupole interactions in the correlator spectrum of optically cooled nuclear spins of a bulk *n*-GaAs sample

© V.M. Litvyak[1], R.V. Cherbunin[1], F.Yu. Soldatenkov[2], V.K. Kalevich[1,2], K.V. Kavokin[1]

[1] Spin Optics Laboratory, St. Petersburg State University,
198504 St. Petersburg, Russia
[2] Ioffe Institute,
194021 St. Petersburg, Russia

E-mail: valiok.ok@gmail.com

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In this work, we measured the correlator spectrum of optically cooled nuclear spin system of a bulk *n*-GaAs crystal in a zero magnetic field. The resulting spectrum is described by two contours, which can be interpreted as the participation of nuclear spins in two types of interactions. We analyzed the measured spectrum in the model for classical magnetic moments in the GaAs lattice. The analysis showed that the main contribution to the high-frequency part of the spectrum is due to the dipole-dipole interaction, and that to the low-frequency part is due to the quadrupole interaction.

Keywords: semiconductors, gallium arsenide, nuclear spin, optical cooling, dipole-dipole interaction, quadrupole interaction, nuclear spin correlator.

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1. Introduction

The magnetic moments of nuclei in the crystal lattice of solids form a thermodynamic system well isolated from the lattice, the spin temperature of which can be several orders of magnitude lower than the absolute value of lattice temperature and have both positive and negative sign [1]. In gallium arsenide, the rates of dipole-dipole and spin-lattice interactions of nuclei can differ by 6 orders [2]. This strong difference allows the nuclear spin system (NSS) temperature to be lowered to a few microkelvin when the crystal itself is at liquid helium temperature. The temperature of NSS in semiconductor crystals can be lowered by optical cooling in a longitudinal magnetic field followed by adiabatic demagnetization into a local field of nuclei. The cooled NSS is able to absorb the power of the alternating magnetic field and warm up in case the frequency of the alternating field coincides with the resonance frequency of the precession of nuclear spins [3,4]. In the absence of an external magnetic field, the resonance frequency of the precession of nuclear spins is determined by the value of the local field. The local field is formed by nuclear spin-spin interactions, which in dielectrics and semiconductors have a magnetic dipole character. The presence of electric field gradients in the studied semiconductor structure leads to quadrupole interactions that increase the spin-spin local field. For strained structures such as quantum wells, quantum dots, and microcavities, quadrupole interactions can increase the local field by an order of magnitude, which has been shown in both experiments and calculations [5–7]. In bulk *n*-GaAs crystals, quadrupole interactions can be related to the presence of residual strain. This leads to an increase in the local field [8], as well as the appearance of an additional peak in the alternating magnetic field power absorption spectra measured at zero magnetic field [9,10].

The ability of cooled NSS to absorb the power of an alternating magnetic field makes it possible to compare experimentally measured thermodynamic and kinetic characteristics of NSS with theoretical calculations using known interaction parameters (for nuclear spins in GaAs — these are the values of dipole-dipole and quadrupole interactions). Among such characteristics are the temporal correlation function and the spectral density of nuclear spin fluctuations. These two functions, defined one on the time axis and the other on the frequency axis, are related by the Fourier transform. In the absence of an external magnetic field they are determined by spin-spin interactions, and for nuclei with spin greater than 1/2 — quadrupolar splitting of nuclear spin levels by local gradients of electric fields. At the same time, the question remains open — how exactly the values of dipole-dipole and quadrupole interactions will influence the type of correlation functions at different values and signs of the nuclear spin temperature. It is assumed that as long as the spin temperature is high enough for the NSS to be in the paramagnetic phase, both of these functions should be independent of the spin temperature. At lowering the spin temperature, a phase transition of nuclear spins into the spin-ordered state is possible, which should be accompanied by a sharp change of both the correlation function and the spectral density of nuclear spin fluctuations (correlator spectrum). It is expected to see an increase in nuclear spin fluctuations when the NSS approaches an ordered

state, and a suppression of fluctuations when nuclear spin order formed. These effects should be manifested in the correlator spectrum and in the associated temporal correlation function of nuclear spins.

In contrast to the formation of ordered states of electron spins possible at liquid helium temperatures (e.g., the formation of a magnetic polaron in semi-magnetic semiconductors [11,12]), the transition of nuclear spins into an ordered state should occur only at spin temperatures of the nanokelvin order caused by the smallness of the nuclear magnetic moment (its value is 3 order of magnitude below the Bohr magneton). This strongly complicates the possibility of experimental observation of the nuclear magnetic order and requires the use of special methods for cooling the NSS. Despite this, in the 1980s years, A. Abragam and M. Goldman observed antiferromagnetic ordering of the nuclear spins of the isotopes ^{19}F in the dielectric CaF_2 [13]. In the late 80s years of the last century, a group of scientists from Rise National Laboratory (Denmark) experimentally observed nuclear spin order in metals, which was achieved at a nuclear spin temperature of about ten nanokelvin [14].

In semiconductor crystals, the ordered state of nuclear spins has not yet been detected. Despite the lack of experimental data, theoretical models for the formation of nuclear spin order in semiconductors are being developed. In 1998, I. Merkulov proposed a model of the nuclear spin polaron, which consists in the formation of a cluster of ordered nuclear spins in the vicinity of the electron localized on the donor center [15]. In this case, the nuclear magnetic order should arise due to the hyperfine interaction between the electron spin and nuclear spins located in the electron localization area and have an antiferromagnetic structure. Since the hyperfine interaction is stronger than the dipole-dipole interaction, the ordering of nuclear spins in the polaron model is possible at higher spin temperatures $\theta_{\mathrm{NP}} \approx 10^{-7}$ K. The polaron model was later developed by D. Scalber in [16], which discusses the detection of the pre-polaron state by a sharp increase in NSS fluctuations. In recent theoretical paper [17], a model of dynamical electron-nuclear ferromagnetism was developed. Such a state, unlike the polaron, is formed at negative spin temperature $\theta_N^d \approx -0.5 \cdot 10^{-6}$ K and possesses long-range order.

Active development of theoretical models of nuclear magnetic order formation in semiconductors awaits experimental confirmation. As noted above, one of the manifestations of nuclear spin ordering is the change in the correlation function of the NSS (nuclear spin correlator) at spin temperatures of the order of fractions of a microkelvin. However, to date, the spectra of nuclear spin correlators and the physical processes involved in their formation are poorly understood even at high spin temperatures.

In this paper, we have measured in zero external magnetic field the frequency spectrum of the NSS correlator of a bulk *n*-GaAs crystal at the nuclear spin temperature of the order of one hundred microkelvin and established the types of inter-nuclear interactions responsible for the shape of the correlator.

2. Experiment procedure

We studied a bulk crystal *n*-GaAs:Si with a donor impurity concentration of $n_d = 1.5 \cdot 10^{16}$ cm^{-3}. The scheme of the experimental setup is given in the paper [10] in Fig. 2. The sample was placed in a closed-cycle cryostat and cooled to a temperature of 6.5 K. The laser diode radiation at a wavelength of 780 nm was passed through a quarter-wave plate ($\lambda/4$), creating a circularly polarized optical pump, and focused on the sample surface. Photoluminescence (PL) was passed through a photo-elastic modulator (PEM), a linear polarizer (GT) and focused on the spectrometer slit. The spectrometer transmitted a PL band at wavelength 817 nm, which was then focused on an avalanche photodiode (APD). The circular polarization of the PL was measured using a two-channel photon counter synchronized with PEM.

The value of the nuclear spin correlator G_ω for a given alternating magnetic field strength can be obtained through a measurement of the heating rate $1/T_\omega$ of the optically cooled NSS.

G_ω is determined by dividing the heating rate as a function of frequency by the square of the AC magnetic field power [1,18]:

$$G_\omega = \frac{4}{\omega^2} \frac{B_L^2}{B_1^2} \left(\frac{1}{T_\omega} \right), \tag{1}$$

where B_L is the local field of the nuclei, B_1 and ω are the amplitude and frequency of the alternating magnetic field, respectively.

We used a technique for measuring the NSS heating spectra of bulk *n*-GaAs crystals first proposed in [18] (see also chapter 12 in [4]) and subsequently developed in our recent [8–10]. A schematic of the experimental protocol and an example curves from which the heating rate $1/T_\omega$ was obtained for a given radio frequency (RF) field strength are given in [10] in Fig. 3.

Our experiment to measure the correlator spectrum consisted of four steps. The stages undergoing on-pumping were monitored by measuring the degree of circular polarization of PL. At first, the NSS was kept in the dark for 10 s in the presence of an alternating magnetic field at 3 kHz in order to erase traces of its cooling acquired during previous measurement cycles (thermalization, Fig. 3, *a* in paper [10]). Next, optical cooling — illumination of the crystal with circularly polarized light in a longitudinal magnetic field $B_z = 150$ G for 60 s followed by adiabatic demagnetization into a local nuclear field B_L was carried out. In result, the nuclear spin system was cooled to a temperature of $\theta_N \approx 7 \cdot 10^{-5}$ K, which is well below the lattice temperature. However, this temperature is not low enough for the formation of an ordered state of nuclear spins in any of the models described above. The third step was exposure of the

cooled NSS to an RF field for a time $t_{rf} = 3$ s in darkness. The amplitude of the alternating magnetic field B_1 was related to the frequency $f = \frac{\omega}{2\pi}$ by the following relation: $B_1 = \frac{kU}{f}$, where U is the voltage applied from the generator to the coil producing the alternating magnetic field, k is the calibration coefficient of the coil. This frequency-amplitude relationship is necessary to maintain the power flux from the RF field in the NSS as the frequency increases. The field frequency for each experiment was taken fixed from the range $f = 10-15 \cdot 10^3$ Hz. The last step was the measuring stage: optical pumping and transverse magnetic field $B_x = 0.5$ G were switched on. In the field B_x, in measure of the remaining spin temperature after heating, a nuclear field arose, the magnitude of which was measured by the change in the degree of circular polarization of the PL. Since the measurement was performed in the $B_x < B_L$ transverse field, which has a magnitude of the order of the Earth's magnetic field, it was necessary to compensate for all three components of the Earth's field. This was done using three pairs of Helmholtz coils.

The treatment of the experimental curves obtained was the same as in paper [10]. First, the heating rate $1/T_\omega$ was calculated from each curve by formula:

$$\frac{1}{T_\omega} = \frac{1}{t_{rf}} \ln\left(\frac{B_N(\omega)}{B_{N0}}\right), \qquad (2)$$

where t_{rf} is the duration of exposure of the NSS to the RF field, $B_N(\omega)$ and B_{N0} are the magnitudes of the nuclear fields after exposure to the RF field and without its exposure, respectively. Then, the spectral density of nuclear spin fluctuations G_ω (correlator spectrum) was calculated using formula (1).

3. Discussion of experimental results

The measured correlator spectrum is represented in Fig. 1 by circles. Each point of the spectrum was obtained at a fixed RF field frequency.

The correlator spectrum consists of two contours: its low-frequency part (up to 1 kHz) is described by the Lorentzian function (dashed line), and the high-frequency part — by the Gaussian function (dash-dotted line). This decomposition can be interpreted as the participation of NSS in two types of interactions. The characteristic time scales of these interactions are related to the half-widths of the Lorentz (500 Hz) and Gauss (2 kHz) contours. As stated above, for GaAs the main type of interactions involving nuclear spins is the magnetic dipole-dipole interaction. It can be described as the effect of the local magnetic field B_{Li} produced by neighboring nuclei on i-th the nuclear spin. Each i-th nuclear spin precesses in such a field with period $T_2 \approx 10^{-4}$ s [2,3]. The magnitude of the scatter of nuclear spin precession frequencies in local fields gives a high-frequency contribution to the correlator spectrum.

In our experiments, the value of the nuclear spin temperature obtained after optical cooling followed by adiabatic

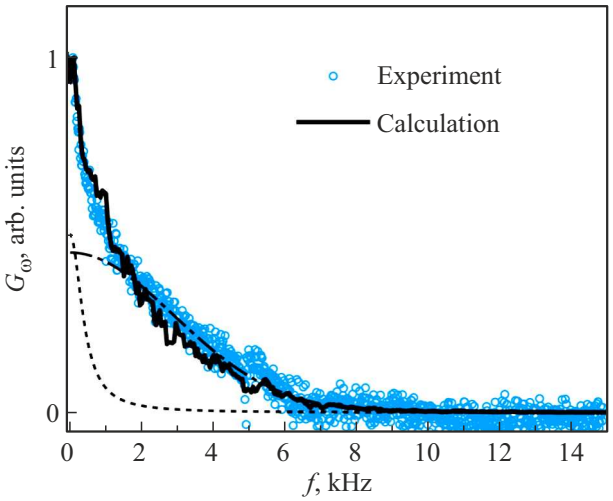

Figure 1. Spectrum of the correlator of cooled nuclear spins of a bulk n-GaAs crystal measured experimentally in zero magnetic field (circles). The spectrum is approximated by the sum of the Lorentzian (dashed line) and Gaussian (dash-dotted line) contours. The solid line shows the spectrum of the nuclear spin correlator calculated in the model for classical magnetic moments in lattice GaAs.

demagnetization turned out to be an order of magnitude higher than the temperature required for the formation of the ordered state of NSS. Therefore, we cannot assume that the low-frequency part of the correlator spectrum we measured is related to the approximation of nuclear spins to magnetic order in any of the existing models [15–17].

Our previous results [9,10] have shown that residual strains leading to quadrupole nuclear spin interactions may be present in bulk n-GaAs crystals. Quadrupole interactions exceeding in magnitude the spin-spin local nuclear field were manifested as a high-frequency absorption peak in the heating spectrum in a zero magnetic field. Therefore, we hypothesized that quadrupole effects caused by uncontrolled residual strain may contribute to the low-frequency part of the correlator spectrum. To test this, we have modelled a correlator for classical magnetic moments in the GaAs lattice. The exact quantum mechanical calculation of the correlator spectrum presents considerable difficulties because of the large number of spins in the lattice, so we used an approximate classical approach based on the solution of the classical equation of motion of magnetic moments. In the GaAs lattice, each nucleus has a mechanical and magnetic moments. Since the mechanical moment of each nucleus precesses in the magnetic field created by neighboring nuclei, its time-dependent direction can be found by solving the N coupled classical equations of motion, where N is the number of spins. Knowing the trajectory of each magnetic moment, we can calculate the total magnetic moment of all nuclei, as well as its correlation function as a function of time. The Fourier transform of the temporal correlation function gives the Fourier image of the correlator, which we can compare with experiment.

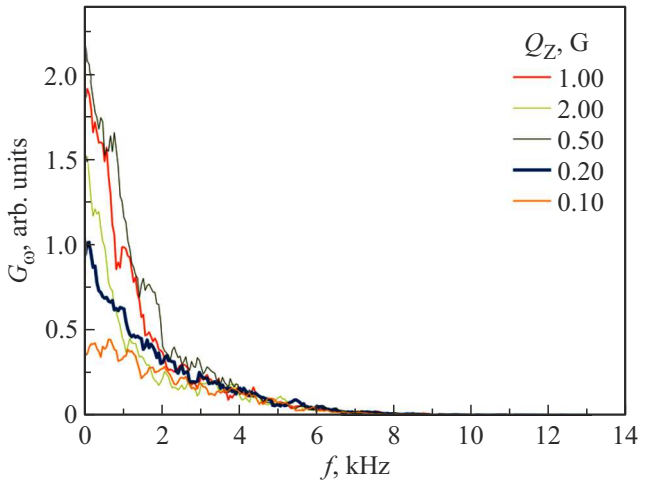

Figure 2. Z-component of the correlator modeled in the classical magnetic moments approximation for different values of the quadrupole interaction Q_Z. The spectrum for the value of the quadrupole interaction equivalent to the splitting of nuclear spin levels in the 0.2 G field (blue curve) best describes the experiment. (A color version of the figure is provided in the online version of the paper).

We considered a small cubic cluster replicating the lattice of gallium arsenide with a natural isotope ratio. The cluster size varied from three to five lattice cells in each direction (approximately from a hundred to a thousand spins). No special conditions were imposed on the spins on the surface of the cluster. Such spins had fewer neighbors and, consequently, lower precession frequencies. In order to determine the role of surface spins, the calculation results were compared with different cluster volumes and, accordingly, with different ratios of the number of spins on the surface and in volume.

At the initial time instant the orientation of the spins was taken random, which corresponds to an infinite spin temperature. At the location of i-th nucleus, the other nuclei create a local magnetic field:

$$B_L^i = \sum_{i \neq j} \frac{3(\mathbf{m}\mathbf{r}_{ij})\mathbf{r}_{ij} - (\mathbf{r}_{ij}\mathbf{r}_{ij})\mathbf{m}}{\mathbf{r}^5}, \qquad (3)$$

where $\mathbf{m}$ is the magnetic moment of the nuclear spin, $\mathbf{r}_{ij}$ is the radius-vector between the i-th and j-th nuclear spins.

This magnetic field causes the direction of i-th magnetic moment to change in time according to the following equation:

$$\frac{d\mathbf{m}_i}{dt} = \gamma_i [\mathbf{m}_i \mathbf{B}_L^i], \qquad (4)$$

where γ_i is the gyromagnetic ratio of the i-th nucleus, to which corresponds the magnetic moment $\mathbf{m}_i$.

Since the local fields change in time, the equation cannot be solved analytically and we found its solution numerically. At each time step, new values of the local fields, magnetic moment directions, and the total moment of the entire cluster $\mathbf{M}(t)$ were calculated. The time step should be smaller than the characteristic precession time of the magnetic moment of the nuclear spin in the local field. This is necessary so that the magnetic moment has time to change its direction at each step. For the isotopes of nuclei in the GaAs crystal, the precession period of each i-th nuclear spin in the effective magnetic field $\mathbf{B}_L^i$ is $T_2 \sim 100\,\mu$s [2]. Therefore, to obtain a more accurate result, the value of the time step in our calculations was taken 2 orders smaller than the time T_2 and was $dt = 1\,\mu$s. The magnitude of the dipole-dipole interaction in the calculation was determined by the constants for the three GaAs crystal isotopes (values of gyromagnetic ratios, radius-vector and magnetic moment values included in formulas (3) and (4)) and gave the same high-frequency part in the modelled correlator spectra (Fig. 2). The fitting parameter was only the magnitude of the quadrupole interaction entered in the calculation in the magnetic field values. The quadrupolar splitting was set uniaxial and directed along a crystallographic axis [100] parallel to the growth axis of the structure. We predict that a possible source of residual strain in the studied structure is the different temperature coefficients of expansion of GaAs and the holder material on which the sample was fixed. Based on our previous papers [9,10], such a source leads predominantly to uniaxial deformation that is directed along the growth axis of the structure. Therefore, we neglected other sources of deformation in this modelling.

Figure 2 shows z-component of the nuclear spin correlator modeled in the classical magnetic moments approximation for different values of uniaxial quadrupole splitting expressed in units of magnetic field. It can be seen that only the low-frequency part of the correlator spectrum changes with its growth. We selected the spectrum that best describes our experiment (the blue spectrum in Fig. 2) from those shown in the figure. This spectrum was modelled for a quadrupole interaction value equivalent to the splitting of nuclear spin levels in a field 0.2 G.

Thus, the calculated curve, taking into account the uniaxial quadrupole interaction equivalent to a magnetic field value of 0.2 G (solid line in Fig. 1), describes the experimentally measured correlator spectrum quite accurately. This result allows to suggest that the low-frequency part of the correlator spectrum is responsible for the quadrupole interaction, the source of which is uncontrolled residual strains in the studied crystal n-GaAs.

4. Conclusion

We have measured the correlator spectrum of the cooled NSS of a bulk n-GaAs crystal with a donor concentration of $n_d = 1.5 \cdot 10^{16}\,\mathrm{cm}^{-3}$. By modelling the correlator for classical magnetic moments in the GaAs lattice, we found that its shape is determined by dipole-dipole and quadrupole interactions. The dipole-dipole interaction is responsible for the high-frequency part of the spectrum, while the

quadrupole interaction is responsible for the low-frequency contribution to the spectrum of the nuclear spin correlator.

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Conflict of interest

The authors declare that they have no conflict of interest.

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