Conference materials
UDC 535
DOI: https://doi.org/10.18721/JPM.161.303

Study of properties of the nuclear spin system in bulk *n*-GaAs by warm-up spectroscopy

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, St. Petersburg, Russia;
[2] Ioffe Institute, St. Petersburg, Russia
✉ valiok.ok@gmail.com

Abstract. In this paper, we present an overview of the possibilities of nuclear spin warm-up spectroscopy method. Nuclear spin warm-up spectroscopy method is based on optical cooling and subsequent warming up of nuclear spins by oscillating magnetic field. Changes of nuclear spin temperature before and after applying of oscillating magnetic field are determined from the degree of photoluminescence polarization. This method is applied to studying the properties of the cooled nuclear spin system in bulk *n*-GaAs crystals. Using warm-up spectroscopy, we can investigate such thermodynamical characteristics of cooled nuclear spins as local fields, absorption coefficients and fluctuations spectral density (correlator spectrum). In particular, such experimental opportunities accompanied by theoretical interpretations allows us to investigate and control the presence of quadrupole interactions in structures. Furthermore, the nuclear spin fluctuations are reflected in the correlator spectrum, which can be recalculated from absorption coefficients. Measurements of the nuclear spin correlator are important experimental opportunity because fluctuations of nuclear spins are one of the main sources of electron spin decoherence in *n*-GaAs.

Keywords: nuclear spin system, optical cooling, absorption spectrum, nuclear spin correlator, local field

Funding: The study was carried out as part of State Assignment “SPbGU” (Topic ID [94030557]), whose funding was used to grow the structures considered. The authors express their gratitude to grant 22-42-09020 from the Russian Science Foundation for funding the experiments.

Citation: Litvyak V.M., Cherbunin R.V., Soldatenkov F.Yu., Kalevich V.K., Kavokin K.V., Study of properties of the nuclear spin system in bulk *n*-GaAs by warm-up spectroscopy, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 (1.3) (2023) 20–25. DOI: https://doi.org/10.18721/JPM.161.303

Материалы конференции
УДК 535
DOI: https://doi.org/10.18721/JPM.161.303

Изучение свойств ядерной спиновой системы объемных слоев n-GaAs методом спектроскопии отогрева

В.М. Литвяк [1] ✉, Р.В. Чербунин [1], Ф.Ю. Солдатенков [2], В.К. Калевич [1,2], К.В. Кавокин [1]

[1] Лаборатория Оптики Спина им. И. Н. Уральцева, СПбГУ, Санкт-Петербург, Россия;
[2] Физико-технический институт им. А.Ф. Иоффе РАН, Санкт-Петербург, Россия
✉ valiok.ok@gmail.com

Аннотация. В данной работе представлены возможности экспериментальной методики спектроскопии отогрева ядерных спинов. Данная методика основана на отогреве переменным магнитным полем оптически охлажденных ядерных спинов. Она позволяет изучать термодинамические характеристики охлажденной спиновой системы ядер объемных слоев *n*-GaAs. К таким характеристикам можно отнести спектры отогрева

и ядерного спинового коррелятора, а также ядерные локальные поля. Создание данной методики направлено на изучение и анализ ядерных спиновых флуктуаций с целью их дальнейшего контроля и подавления.

Ключевые слова: ядерная спиновая система, оптическое охлаждение, спектр поглощения, ядерный спиновый коррелятор, локальное поле

Финансирование: Работа выполнена в рамках Государственного задания «СПбГУ» (код темы [94030557]), с поддержкой которого были выращены изучаемые структуры. Авторы также выражают благодарность гранту РНФ No 22-42-09020 за финансовую поддержку в проведении экспериментальных исследований.

Ссылка при цитировании: Литвяк В.М., Чербунин Р.В., Солдатенков Ф.Ю., Калевич В.К., Кавокин К.В. Изучение свойств ядерной спиновой системы объемных слоев *n*-GaAs методом спектроскопии отогрева // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2023. Т. 16. № 1.3. С. 20–25. DOI: https://doi.org/10.18721/ JPM.161.303

Introduction

One of the main tasks of modern spintronics is the realization of the possibility of using the electron spin as an information carrier. However, the main obstacle to this is the electron spin relaxation. We need to look for ways to increase the electron spin lifetime. In bulk *n*-GaAs structures, the maximum of an electron lifetime localized on a donor center is observed at a donor impurity concentration near the metal-insulator transition [1]. Therefore, it is promising to use *n*-GaAs samples with a donor concentration of the order of $n_D = 10^{15}$–10^{16} cm^{-3}. Wherein in such structures, one of the main mechanisms of electron spin relaxation is the interaction with nuclear spin fluctuations [1, 2]. To increase the electron spin lifetime, one must be able to control and, ideally, suppress the nuclear spin fluctuations.

Suppression of nuclear spin fluctuations is possible at ultralow nuclear spin temperatures, on the order of fractions of a microkelvin. At such temperatures, a transition of nuclear spins to a magnetic order is expected. In 1997, Merkulov proposed the theoretical concept of a nuclear spin polaron in bulk *n*-GaAs [3]. According to Merkulov, when the nuclear spin system (NSS) is cooled to temperatures of the order of fractions of a microkelvin, nuclear spins are expected to pass into an ordered state in the vicinity of the donor center, due to hyperfine interaction with the electron. To date, there have been published several theoretical works about on the nuclear spin ordering due to hyperfine interaction in semiconductors [4–6]. But none of these ordered states has yet been observed experimentally. However, this is one of the main possibilities for suppressing nuclear spin fluctuations. The detection of the pre-polaron state is supposed to be based on the change in the low-frequency part of the nuclear spin fluctuations spectrum (correlator spectrum) [3, 6]. It is expected that as the nuclear spin system (NSS) approaches to the ordered state, the low-frequency part of the correlator spectrum should increase its magnitude. To observe this effect, it is necessary to implement two conditions of the experiment. It is necessary to be able to cool the NSS to microkelvin spin temperatures and to reliably measure the nuclear spin correlator. The nuclear spin warm-up spectroscopy method, proposed in this paper, allows one to carry out experiments on deep cooling of the NSS in *n*-GaAs. Also, using this method, we can obtain the correlator spectrum of optically cooled nuclear spins by measuring the absorption coefficients at different frequencies of an oscillating magnetic field (OMF).

In this paper, we present the results obtained with nuclear spin warm-up spectroscopy method for bulk *n*-GaAs samples. The basics of this method were laid in early works of 1980s [7, 8], where first absorption spectra of the NSS in a bulk *n*-GaAs sample were measured, but the physical mechanisms of formation of the obtained spectra were not elucidated. To date, we have come closer to understanding the physical foundations of absorption spectra. In particular, it turned out that even otherwise insignificant quadrupole effects have a strong influence on their shape

[9, 10]. We have not yet fully explored the mechanisms which determine the nuclear spin correlator spectra. However, the correlators measured by our method are qualitatively consistent with the shapes of spin correlation functions, obtained earlier by numerical modeling [11, 12]. This gives us confirmation that our nuclear spin warm-up spectroscopy method really makes it possible to detect nuclear spin fluctuations. It means that the development of this method together with a theoretical analysis will eventually allow us to learn how to detect an approach of the NSS to the polaron state.

Materials and Methods

With the help of nuclear spin warm-up spectroscopy method, the NSS of bulk *n*-GaAs layers with donor impurity concentrations in the range of 10^{15}–10^{16} cm^{-3} was studied. The samples under studies were grown by liquid-phase epitaxy. The scheme of the experimental setup is given in [9]. The sample under study was placed in a closed cycle cryostat and cooled down to 6 K. Radiation from a laser diode at a wavelength of 780 nm passed through a quarter-wave plate, creating a circularly polarized optical pump, and was focused on the sample surface. The polarized photoluminescence (PL) signal passed through a photoelastic modulator (PEM) and a linear polarizer and was focused on the slit of the spectrometer. The spectrometer passed the PL line at a wavelength of 817–819 nm (depending on the sample under study), which then was focused on the photodiode chip. The PL intensity was determined using a two-channel photon counter connected to a photodiode and synchronized with the PEM. All measurements were carried out by detecting the change in the degree of PL polarization with time.

As some examples of experimental results, the measured absorption spectra in a zero magnetic field, the correlator spectrum, and the magnitude of the local field will be given in this work. To obtain these characteristics, we used a multi-stage experimental protocol proposed in [7, 8] and developed in [9, 10].

The experimental protocol used to obtain the absorption spectrum in zero magnetic field consisted of four stages. The first stage is called preparatory. The NSS was in the dark during the minute. By the end of this stage, the NSS came to thermodynamically equilibrium state. The next stage was optical cooling of the NSS. For a minute, the sample was pumped with circularly polarized light in a longitudinal magnetic field B_Z = 150 G. Spin-polarized electrons polarized nuclear spins due to hyperfine interaction. By the end of the second stage, a nuclear field and the corresponding spin temperature, different from the lattice temperature, were created. This was followed by adiabatic demagnetization to zero external field. The longitudinal field B_Z was switched off to zero during a time is equal to 20 ms. After demagnetization, the spin temperature decreased by several orders of magnitude. For structures under study at the described experimental conditions the nuclear spin temperature was about 100 μK. The third stage was the application of an OMF in the dark for 3 s at a fixed frequency. To obtain the absorption spectrum in a zero static magnetic field, the frequencies were chosen from the range from 100 Hz to 20 kHz at a fixed amplitude of OMF. After the applying of OMF, the nuclear spin temperature was increased. The heating process took place at the rate of $(1/T_\omega + 1/T_1)^{-1}$, where $1/T_\omega$ is the heating rate due to the impact of OMF on the NSS at a frequency ω, $1/T_1$ is the relaxation rate of the NSS due to spin-lattice relaxation. The last step was measuring the nuclear field B_N, which remain after OMF application. To do this, the pump and the transverse magnetic field B_X (measurement field) were switched on. Electron spins depolarized in the total field $B_X + B_N$, and then during the whole measuring stage (about 200 s) the nuclear field B_N decreased due to spin-lattice relaxation, and the electron polarization was restored. An example of the protocol described above is given in [9] in Fig. 3.

The rate of the NSS heating is related to the magnitude of the nuclear field B_N remaining after the application of OMF at frequency ω by the following formula: $1/T_\omega = (1/t_{OMF})\cdot\ln[B_N(\omega)/B_{N0}]$, where B_{N0} is the value of the nuclear field in the case, when in the third stage the OMF was not applied. Ref. [9] describes in detail how to extract the absorption coefficient $1/T_\omega$ from the fitting of a multi-stage curve.

Spectral fluctuation density G_ω (nuclear spin correlator) is related to the heating rate by the following formula: $G_\omega = (1/T_\omega)\cdot(4B_L^2/\omega^2 B_1^2)$ [7], where B_L is the local field, B_1 is the amplitude of OMF, ω is the frequency of OMF. Therefore, the experimental protocol for obtaining the correlator spectrum remains the same. The only difference is that the measurement takes place in a narrower and lower frequency range of OMF frequencies: from 10 Hz to 15 kHz.

To measure the magnitude of the local field B_L, the stage of an OMF application is not needed. Therefore, only three stages remain from the protocol described above: thermalization, optical cooling with adiabatic demagnetization and measurement. In order to determine the value of the local field, we proceeded to the last stage, carrying out measurements with the magnitude of B_X varied from 0.2 G to 7 G at a step from 0.1 to 1 G, depending on steepness of the dependence $B_N(B_X)$. Further, from each measurement the nuclear field B_N was extracted, and compared with the theoretical dependence:

$$B_N = b_N \hbar \langle \gamma_N \rangle \frac{I(I+1)}{3k_B} B_X \beta \sqrt{\frac{B_L^2}{B_L^2 + B_X^2}}, \quad (1)$$

where b_N is the Overhauser field at full nuclear polarization, k_B is the Boltzmann constant, $\hbar$ is the Planck constant, $\langle \gamma_N \rangle$ is the average gyromagnetic ratio of the NSS of GaAs, $I = 3/2$ is the nuclear spin, β is the inverse nuclear spin temperature. The obtained dependence $B_N(B_X)$ was fitted by Eq. (1) with the value of B_L as a free parameter. In this way, the value of the local field was determined.

Results and Discussion

In this section, examples of absorption spectra at zero external magnetic field, correlator spectrum and experiment for determination of the local field are presented. These results demonstrate the experimental possibilities of the nuclear spin warm-up spectroscopy method. Also, it should be noted, that for results, presented below, the value of the nuclear spin temperature that was reached by the end of the second stage of multistage measurements (after adiabatic demagnetization) was several orders of magnitude higher than the temperature required to achieve the polaron state. Obtaining ultralow nuclear spin temperatures is a separate experimental direction, not included in this paper.

Absorption spectra for the sample with the donor concentration $n_D = 5 \cdot 10^{15}$ cm^{-3} are shown in Fig. 1. These spectra were measured in three different points on the sample surface (Point *1*, Point *2*, Point *3*). The main difference between spectra is the frequency positions of the absorption peaks - they depend on the magnitude of the quadrupole interaction of nuclear spins. So, at the Point *1* (dots in Fig. 1) the magnitude of the quadrupole interaction is considered to be minimal, and at the Point *2* (triangles in Fig. 1) the magnitude of the quadrupole interaction is the largest. In this case, two absorption peaks at frequencies of the order of 4 kHz and 9 kHz (Point *2*, Point *3*) correspond to the precession of isotopes ^{75}As and ^{69}Ga together with ^{71}Ga in the local fields, determined by dipole-dipole and quadrupole interactions. These frequencies can vary depending on the magnitude of the quadrupole splitting [9, 10]. The experimental fact that in different points on the sample surface the different absorption spectra are obtained indicates the spatial inhomogeneity of the quadrupole interaction of nucleus in the sample, which can be revealed by our nuclear spin warm-up spectroscopy method.

The example of nuclear spin correlator spectrum, which was obtained for the sample with donor concentration $n_D = 1.2 \cdot 10^{16}$ cm^{-3}, is shown in Figure 2 by dots. This spectrum corresponds to the nuclear spin temperature of about 50 μK (optical cooling stage time was equal to 60). The low-frequency part of the correlator spectrum is described by a Lorentzian (dashed line), and high-frequency part is described by a Gaussian (dash-dotted line). Such a decomposition of the spectrum into contours is in agreement with the temporal spin correlation functions calculated within different models [11, 12], which are related to the frequency dependence through the Fourier transform. It should be noted that the shape of the nuclear spin correlator spectra is the same for all studied bulk *n*-GaAs crystals with donor concentrations in the range of 10^{15}–10^{16} cm^{-3}. This indicates, in particular, that the donor

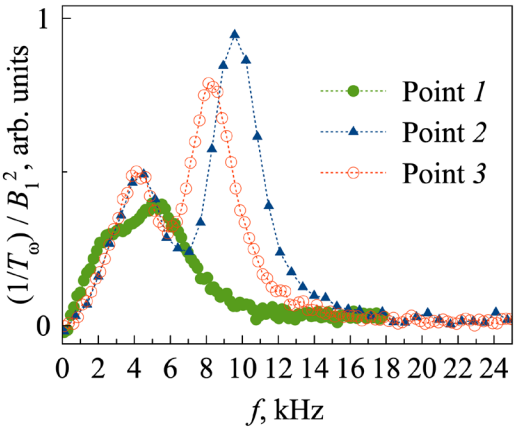

Fig. 1. Absorption spectra in three different points on the sample surface (Point *1*, Point *2*, Point *3*) at zero external magnetic field

concentration does not affect the formation of the spectra of nuclear spin fluctuations. In Fig. 3, the black dots show the dependence of the value of nuclear field B_N on the amplitude of the measuring field B_X, constructed according to the method described in the previous section. The obtained experimental dependence was described by Eq. (1) with the value of the local field B_L as a fitting parameter. For the sample under study, the value of local field is equal to $B_L = (1\pm0.1)$ G, which is close to the known literature data for bulk GaAs [13].

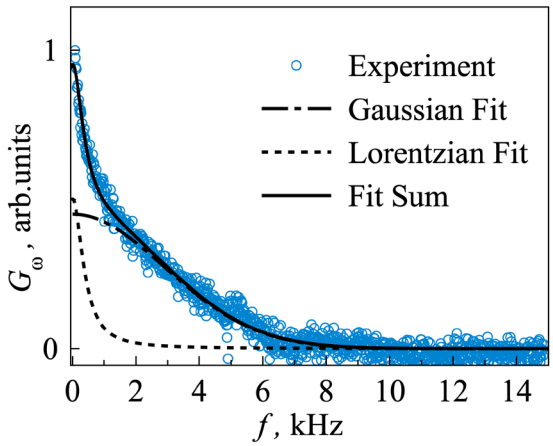

Fig. 2. Nuclear spin correlator spectrum for bulk n-GaAs (dots).
The spectrum was obtained by the warm-up spectroscopy method. The parts with low and high frequencies are described by Lorentzian and Gaussian functions, respectively

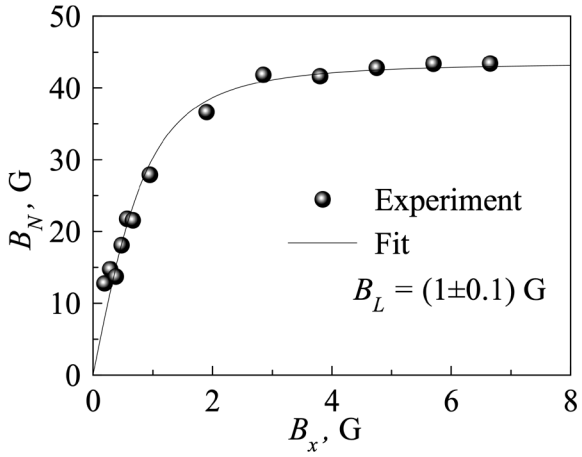

Fig. 3. Experimental dependence of nuclear field on the measurement transverse magnetic field for determination of the local field (black dots). Fitting experimental results by Eq. (1) with B_L as fitting parameter (black curve)

Conclusion

In this work, the experimental possibilities of nuclear spin warm-up spectroscopy method were presented for bulk n-GaAs samples with donor concentrations in the range of 10^{15}–10^{16} cm^{-3}. The proposed method makes it possible to obtain such characteristics of a cooled NSS as absorption and correlator spectra, and also makes it possible to measure nuclear local fields. In particular, we found that the absorption spectrum in a zero magnetic field reflects the spatial inhomogeneity of the quadrupole interaction, the correlator spectra consist of two contours, and their frequency shape does not depend on the donor concentration.

REFERENCES

1. **Dzhioev R. I., Kavokin K. V., Korenev V. L., Lazarev M. V., Meltser B. Ya., Stepanova M. N., Zakharchenya B. P., Gammon D., Katzer D. S.** Low-temperature spin relaxation in n-type GaAs, Physical Review B. 66 (2002) 245204.

2. **Merkulov I. A., Efros Al. L., Rosen M.** Electron spin relaxation by nuclei in semiconductor quantum dots, Physical Review B. 65 (2002) 205309.

3. **Merkulov I. A.** Formation of a nuclear spin polaron under optical orientation in GaAs-type semiconductors, Physics of the Solid State. 40 (1998) 930–933.

4. **Scalbert D.** Nuclear polaron beyond the mean-field approximation, Physical Review B. 95 (2017) 245209.

5. **Fischer A., Kleinjohann I., Anders F, Glazov M. M.** Kinetic approach to nuclear-spin polaron formation, Physical Review B. 102 (2020) 165309.

6. **Vladimirova M., Scalbert D., Kuznetsova M. S., Kavokin K. V.** Electron-induced nuclear magnetic odering in n-type semiconductors, Physical Review B. 103 (2021) 205207.

7. **Kalevich V. K., Fleisher V. G.** Optical detection of NMR with dynamic cooling of the nuclear spin system of a semiconductor by polarized light, Bull. Acad. Sci. USSR, Phys. Ser. 47 (1983) 5–9.

8. **Kalevich V. K., Kulkov V. D., Fleisher V. G.** Onset of a nuclear polarization front due to optical spin orientation in a semiconductor, JETP Lett. 35 (1982) 17–20.

9. **Litvyak V. M., Cherbunin R. V., Kalevich V. K., Lihachev A. I., Nashchekin A. V., Vladimirova M. and Kavokin, K.V.** Warm-up spectroscopy of quadrupole-split nuclear spins in n-GaAs epitaxial layers, Physical Review B. 104 (2021) 235201.

10. **Litvyak V. M., Cherbunin R. V., Kavokin K. V. and Kalevich V. K.** Determination of the Quadrupole Splitting in Bulk n - GaAs by Warm - Up Spectroscopy, Semiconductors. 54 (2020) 1728–1729.

11. **Knak Jensen S. J., Kjaersgaard Hansen E.** Dynamics of classical spins with dipolar coupling in a rigid lattice at high temperatures, Physical Review B. 13 (1976) 1903–1918.

12. **Florencio J., O. F. de Alcantara Bonfim.** Recent Advances in the Calculation of Dynamical Correlation Functions, Frontiers in Physic. 8 (2020) 1–13.

13. **Paget D., Lampel G., Sapoval B., Safarov V. I.** Low field electron-nuclear spin coupling in gallium arsenide under optical pumping conditions, Physical Review B. 15 (1977) 5780.

THE AUTHORS

LITVYAK Valentina M.
valiok.ok@gmail.com
ORCID: 0000-0001-9142-6821

CHERBUNIN Roman V.
r.cherbunin@gmail.com
ORCID: 0000-0002-8791-6227

SOLDATENKOV Fedor Yu.
f.soldatenkov@mail.ioffe.ru
ORCID: 0000-0002-6154-7208

KALEVICH Vladimir K.
kalevich@solid.ioffe.ru
ORCID: 0000-0002-8010-2714

KAVOKIN Kirill V.
kkavokin@gmail.com
ORCID: 0000-0002-0047-5706

Received 29.11.2022. Approved after reviewing 23.01.2023. Accepted 25.01.2023.