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Absorption of an oscillating magnetic field power at frequencies of nuclear spin-spin interactions in semiconductors

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We present a general overview of our previously published experimental and theoretical studies of the power absorption spectra of oscillating magnetic fields by optically cooled nuclear spins in semiconductors of the A^3B^5 and A^2B^6 groups. Absorption spectra were measured in zero and also in weak external static magnetic fields using warm-up spectroscopy, which is an analogue of optically detected nuclear magnetic resonance (ODNMR). The experiments were carried out for a deformed bulk *n*-GaAs layer and a CdTe-based heterostructure. It is shown that the shape of the absorption spectra is determined by nuclear spin-spin interactions, which are very different for the selected semiconductor materials. In particular, from a theoretical analysis of experimental spectra it was established that for an *n*-GaAs crystal in the presence of residual deformation, the shape of the absorption spectrum in a zero magnetic field is determined by quadrupole interactions exceeding nuclear spin-spin interactions. For the CdTe heterostructure, it was established that the shape of the spectrum in zero field has a purely spin-spin character, which is confirmed by our proposed model of nuclear spin clusters. For both *n*-GaAs, and CdTe, absorption spectra were measured and analyzed in external static magnetic fields (analogue of NMR), where homonuclear (Zeeman) absorption peaks were found to be modified by the presence of a nuclear quadrupole (for *n*-GaAs) and pure spin-spin (for CdTe) interactions.

Keywords: semiconductors, nuclear spin, optical cooling, absorption spectrum, spin-spin interactions.

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1. Introduction

In most A^3B^5 and A^2B^6 semiconductors nuclear isotopes have non-zero spin moments, which determine the macroscopic nuclear magnetization. One of the ways to create a macroscopic nuclear magnetic moment is the optical cooling of nuclear spins by spin-polarized electrons in an external magnetic field [1]. This method of creating nuclear spin polarization, which corresponds to the nuclear field B_N (Overhauser field), is possible due to the hyperfine interaction of nuclear and optically oriented electron spins. The magnetization created in this way can follow the predictions of nuclear spin temperature theory for an adiabatic change in the external magnetic field through zero field [2]. This process is called adiabatic demagnetization and is used for deep cooling of the nuclear spin system (NSS) [3,4]. The nuclear spin temperatures before (θ_N) and after (θ_N^{ad}) the adiabatic demagnetization are related by the expression

$$\theta_N^{ad}/\theta_N = \sqrt{B_L^2/(B_L^2 + B_Z^2)}, \qquad (1)$$

where B_Z — the external longitudinal (relative to the pumping beam) magnetic field in which optical cooling was done.

Formula (1) shows that demagnetization occurs from the external longitudinal field B_Z to the nuclear local field B_L. This important point suggests that the local field determines the minimum spin temperature that can be achieved in optical cooling experiments followed by adiabatic demagnetization. It is noteworthy that achieving ultra-low nuclear spin temperatures in semiconductors is one of the current experimental tasks on the way to implementing existing concepts of the transition of nuclear spins to a magnetic-ordered state [5–7]. Therefore, it is important to understand the physical processes involved in the formation of the nuclear local field in the semiconductor structure under study.

The local field in semiconductors is determined by nuclear spin-spin interactions. These include long-range magnetic dipole-dipole interaction, as well as indirect interactions through valence electrons (exchange and pseudo-dipole) [8]. Indirect interactions are short-range and are taken into account only for nearest neighbors. If nuclear isotopes have spin $I > 1/2$, then they can participate in quadrupole interactions, which increase the pure spin-spin local field [9,10]. Thus, the magnitude of the nuclear local field has two main contributions: spin-spin (B_{SS}) and quadrupole (B_Q):

$$B_L = \sqrt{B_{SS}^2 + B_Q^2}. \qquad (2)$$

There exists a direct measuring method of the magnitude of the local field B_L, based on the adiabatic demagnetization of nuclear spins. Using this method, we previously

determined the local field for an *n*-GaAs crystal with a minimum residual strain [8]. However, it is important to understand how all the available contributions to the magnitude of the local field appear in NMR and ODNMR experiments, in which the effects associated with nuclear magnetization most clearly appear.

We can highlight a number of fundamental experimental and theoretical works devoted to a detailed study of the influence of nuclear spin-spin interactions on the shape of NMR spectra in strong magnetic fields for A^3B^5 semiconductors, in particular for GaAs [11–13]. All isotopes in GaAs are magnetic and have spin $I = 3/2$. These features of the NSS give rise to broadened absorption peaks, which, in the presence of deformation in the crystal, are accompanied by quadrupole satellites. For a CdTe crystal, in which the available magnetic isotopes (their abundance is weak compared to GaAs and is about 25%) have spin $I = 1/2$ and do not participate in quadrupole interactions, the shape of the NMR spectrum is determined by Zeeman, as well as purely spin-spin interactions. This was shown in the work by Nolle in 1979 [14]. The Zeeman absorption peak of the isotope Te^{125}, measured in a strong magnetic field, was accompanied by purely spin-spin satellites.

Based on the differences in the spin properties of semiconductor structures of *n*-GaAs and CdTe described above, we previously carried out a series of experiments devoted to measuring absorption spectra in zero and weak external magnetic fields in order to study how spin-spin interactions affect the shape of such spectra. To analyze our experimentally measured spectra, theoretical models were constructed that allow us to identify the observed absorption peaks, as well as make up conclusions about the contributions of nuclear spin-spin interactions to their shape. We obtained all the experimental and theoretical results presented below earlier and published them in the articles [8,9,15–21]. This work is of a review nature and allows to take a comprehensive look at the main results of our previous works.

2. Samples and experimental procedure

This work will present the experimentally measured warm-up spectra upon absorption of the power of an alternating magnetic field, as well as their theoretical analysis for two semiconductor structures: a bulk deformed layer of *n*-GaAs and a heterostructure based on CdTe. There will be an overview of the main results we have previously obtained from the point of view of their comparative analysis. Such a review is important for a comprehensive view at the currently known features of nuclear spin-spin interactions in the studied structures, which appear in the warm-up spectra.

The first structure was a bulk GaAs layer of $77\,\mu$m thickness, doped with silicon and grown on a substrate *p*-GaAs [001] by liquid-phase epitaxy. The donor impurity concentration was $n_d \approx 10^{15}\,\mathrm{cm}^{-3}$. Details of growth are given in the work [15]. The main experimental results for this sample are presented in our previous works [16,17]. In particular, it was found that this structure contains residual deformations that exceed the energy of nuclear spin-spin interactions. Deformations lead to the emergence of nuclear quadrupole interactions, which determine the shape of the warm-up spectra upon absorption of the power of an alternating magnetic field both in zero and in external static magnetic fields.

The second structure was an undoped single quantum well (QW) of CdTe, $d = 30\,$nm wide, separated from the buffer layer by a $Cd_{0.95}Zn_{0.05}Te$ barrier with a thickness of 1064 nm, and from the surface by a $Cd_{0.95}Zn_{0.05}Te$ barrier, 93 nm thick. The heterostructure under study was grown by molecular-beam epitaxy on a $Cd_{0.96}Zn_{0.04}Te$ substrate [100] . The results of studying the optical properties and nuclear spin dynamics for this heterostructure are presented in the works [18–19,21–22]. The main important feature of the CdTe semiconductor structure from the point of view of the nuclear spin system is the absence of quadrupole interactions, as well as the low abundance of magnetic isotopes.

The warm-up spectra, which will be discussed below, were measured in the framework of our previously developed technique for warm-up spectroscopy of optically cooled nuclear spins [16,17]. This method allows one to study the thermodynamic characteristics of a cooled NSS, which, in addition to the warm-up spectra in zero and external static magnetic fields (an analogue of ODNMR), can include the spectra of the nuclear spin correlator, as well as nuclear local fields. It is noteworthy that the ability to measure in a reliable way and analyze theoretically these characteristics of NSS allows us to expand the understanding of the features of the state of a cooled NSS for a specific semiconductor structure which is under study. A general review of the experimental capabilities of the nuclear spin warm-up spectroscopy technique was published in our recent work [20].

The diagram of the experimental setup using this method is shown in Figure 2 in the work [16]. The sample under study was placed in a closed-cycle cryostat and cooled to a temperature of $7-12\,$K. The laser diode radiation at a wavelength of 780 nm (for GaAs) and at a wavelength of 680 nm (for CdTe) passed through a quarter-wave plate ($\lambda/4$), creating a circularly polarized optical pump, and was focused on the surface sample. Measurements of the absorption coefficient at a fixed frequency of an alternating magnetic field were carried out by detecting changes in the degree of polarization of photoluminescence (PL) with time according to a given optical protocol. The spectrometer transmitted the PL band at a certain wavelength (817 nm for GaAs and 774 nm for CdTe), then the PL was focused on an avalanche photodiode (APD).

To measure the warm-up spectra of the *n*-GaAs sample, a four-stage optical protocol was used, described in detail in [16]. The measurement of absorption coefficients is based on the method of optical cooling of the NSS for

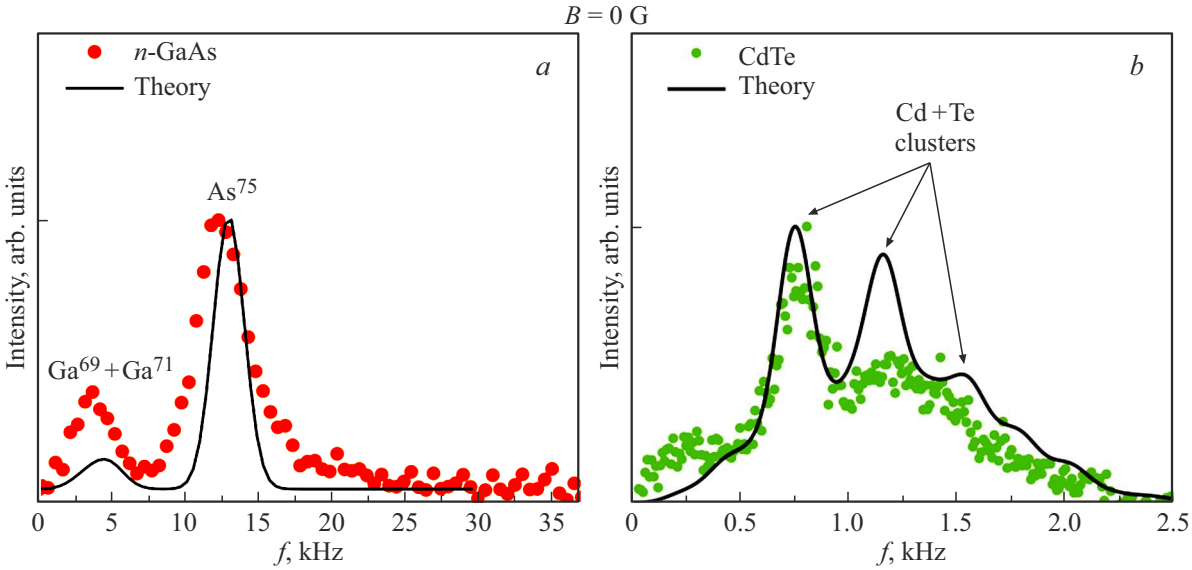

Figure 1. Warm-up spectra upon absorption of the power of an alternating magnetic field, measured in a zero static magnetic field for (a) bulk deformed layer n-GaAs (red dots) and for (b) CdTe QW (green dots). The spectra obtained from the simulation are shown in black lines.

several minutes in a longitudinal magnetic field, followed by adiabatic demagnetization into a local field B_L. This created nuclear magnetization, which corresponds to an inverse nuclear spin temperature β, several orders of magnitude higher than the lattice reciprocal temperature, as well as a nuclear field B_N. Next, an alternating magnetic field was applied to the cooled NSS for a time t_{OMF} at a fixed frequency f. If the frequency f is close to the resonant frequency of the nuclei, there was warm-up of the NSS: β and $B_N(f)$ decreased. To detect the warm-up rate $(1/T_{\mathrm{OMF}})$, a weak measuring field B_x was turned on. The electron spins were depolarized in the total field $B_x + B_N$, while the nuclear field B_N at the first moment of time of the measuring stage was formed to the extent of the inverse spin temperature β achieved at the previous stage according to the expression

$$B_N = \left(I(I+1)b_N\hbar\bar{\gamma}_N/3k_b\right)B_x\beta\sqrt{B_L^2/(B_L^2+B_x^2)}, \qquad (3)$$

where b_N — Overhauser field at 100% nuclear polarization, k_b — Boltzmann's constant, $\hbar$ — Planck's constant, $\bar{\gamma}_N$ — isotope-averaged nuclear gyromagnetic ratio, I — nuclear spin.

The warm-up rate for each frequency f was recalculated using the formula

$$1/T_{\mathrm{OMF}} = (1/t_{\mathrm{OMF}})\ln\left(B_{N0}/B_N(f)\right), \qquad (4)$$

where B_{N0} — is the nuclear field in the absence of an alternating magnetic field.

The result of these measurements is the warm-up spectrum: dependence of $1/T_{\mathrm{OMF}}$ on f. If we talk about warm-up spectra in external magnetic fields, then at the stage of exposure to an alternating magnetic field, an additional static magnetic field was applied across the direction of the alternating one (analogous to NMR).

To measure the warm-up spectra of the CdTe heterostructure, a similar optical protocol was used, but for effective nuclear polarization, the optical cooling time was increased to 700 seconds. This protocol is described in detail in the works [18,19].

3. Results and discussions

This part of the work will present experimentally measured warm-up spectra for a deformed bulk layer n-GaAs and for a CdTe heterostructure, as well as theoretical models that explain the observed spectral features. In particular, the warm-up spectra of optically cooled nuclear spins in a zero static magnetic field, as well as in an external static magnetic field $B_x = 14.5\,\mathrm{G}$ will be analyzed. For n-GaAs a spectrum will be presented measured in a sufficiently strong magnetic field ($B_x = 48.3\,\mathrm{G}$) with the aim of showing the appearance of quadrupole interactions caused by mechanical deformation of the lattice.

In our early studies for a bulk deformed layer n-GaAs [16,17] it was established that quadrupole effects manifest themselves in the warm-up spectrum in a zero field as an additional high-frequency peak. Meanwhile, the frequency position of this peak depends on the magnitude of the deformation. An example of such a warm-up spectrum is shown in Figure 1, a with red dots. The spectrum contains two broad peaks: low-frequency ($f_1 \sim 3.5\,\mathrm{kHz}$) and high-frequency ($f_1 \sim 12\,\mathrm{kHz}$) peaks with half-width at half-maximum (HWHM) $\sim 3.5\,\mathrm{kHz}$. We interpreted the positions of these peaks as characteristic precession frequencies of the isotopes As^{75} and $Ga^{69.71}$ in local fields determined

Spectral features of the measured warm-up spectra in a zero magnetic field for deformed *n*-GaAs and CdTe

Semiconductor structure	Time of optical cooling, s	HWHM, kHz	f_n, kHz
n-GaAs (deformed)	60–120	3.5	$f_1 \sim 3.5$ $f_2 \sim 12$
QWCdTe	700	0.2	$f_1 \sim 0.78$ $f_2 \sim 1.2$ $f_3 \sim 1.5$

by spin-spin and quadrupole interactions. It is noteworthy that the warm-up spectrum for a similar bulk layer *n*-GaAs, but with a minimum value of residual deformation, contains one peak at the frequency $f_1 \sim 3.5$ kHz (red spectrum in Figure 1 in [8]) with HWHM ~ 3.5 kHz. The observed single peak indicates that quadrupole interactions in the sample have an energy comparable to the energy of purely spin-spin interactions of nuclei.

In the warm-up spectrum in a zero static magnetic field for a CdTe heterostructure, it was expected to obtain one narrow low-frequency peak associated with the precession of nuclear spins in local fields formed by purely spin-spin interactions in the absence of quadrupole effects. However, the measured spectrum has a more complex shape (Figure 1, *b*, green dots). It is formed in the low-frequency spectral region (up to 2.5 kHz) and instead of one peak contains at least three narrow low-frequency peaks ($f_1 \sim 0.78$ kHz, $f_2 \sim 1.2$ kHz and $f_3 \sim 1.5$ kHz) with HWHM ~ 0.2 kHz and a high-freqency tail. We assumed (and then confirmed by a theoretical model) that the nature of the observed absorption peaks is associated with the peculiarities of spin-spin interactions in CdTe: it is known that in this material only approximately 25% of all isotopes have non-zero magnetic moments, in contrast to GaAs, and also the constants of nuclear spin-spin interactions are different. These properties, in particular, occurred in the warm-up spectrum in a zero magnetic field. The main constants characterizing the NSS in GaAs and CdTe are given in the table in the work [18].

Spectral features for the measured warm-up spectra in a zero magnetic field are given in the table. The table also shows the times during which optical cooling was conducted. To achieve noticeable nuclear magnetization in CdTe, the cooling time was increased by an order of magnitude compared to GaAs, which is due to the low abundance of magnetic isotopes.

The quadrupole nature of the high-frequency warm-up peak f_2 for the deformed layer *n*-GaAs was confirmed in our early work [16], where in addition to the warm-up spectrum in zero field, a series of spectra were measured in external magnetic fields in four mutual orientations of alternating and static magnetic fields. The results of the experiments are shown in the work [16] in Figure 6 with colored dots. The measured spectra were calculated as part

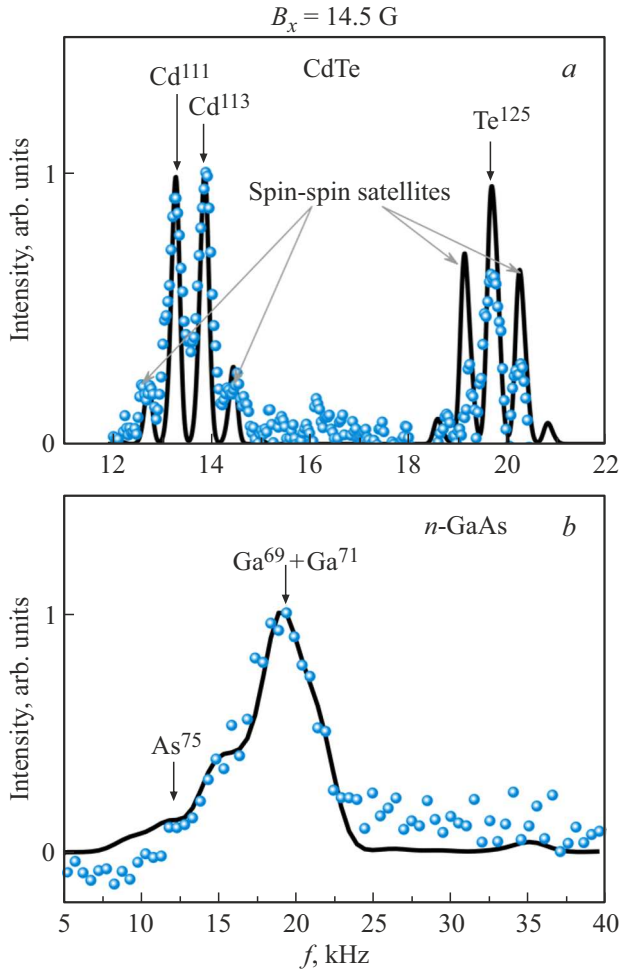

Figure 2. Examples of measured warm-up spectra in an external field $B_x = 14.5$ G for a CdTe (*a*) heterostructure and for a deformed bulk layer *n*-GaAs (*b*) are shown with blue dots. The spectra obtained from the simulation are shown in black lines. Grey and black arrows indicate spin-spin satellite and homonuclear absorption peaks, respectively.

of a model that took into account various potential sources of quadrupole interactions, the values of which were fitting parameters. From fitting the calculated spectra to the measured ones, it was specified that the source of quadrupole interactions in the bulk *n*-GaAs layer is mechanical stress. In Figure 1, *a* the experimental warm-up spectrum (red dots) is described by a spectrum modeled taking into account quadrupole interactions, which is represented by a black line. It is noteworthy that when simulating, the half-width at half-maximum of the absorption lines was taken equal to HWHM = 1.5 kHz. A detailed description of the theoretical model is also given in the work [16].

To explain the warm-up spectrum of CdTe QW measured in a zero magnetic field, we proposed a model of non-interacting nuclear spin clusters consisting of different numbers of spins. Details of the model are presented in the work [18]. In particular, clusters consisting of 2, 3, 4 and 5 isotopes of CdTe were reviewed. It turned out that the

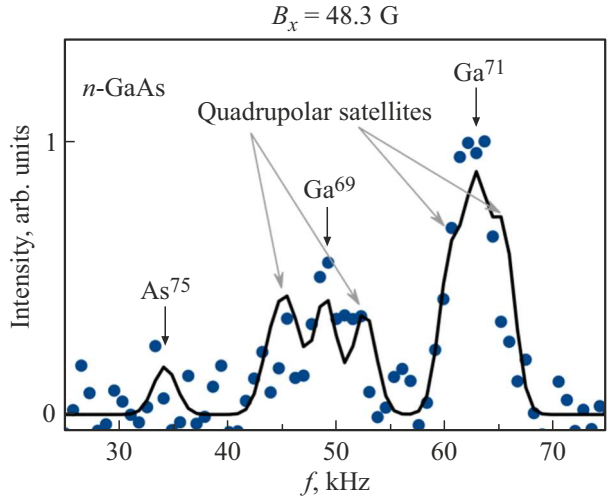

Figure 3. The examples of the measured warm-up spectrum in an external field $B_x = 48.3$ G for a deformed bulk layer *n*-GaAs (blue dots). Its spectrum, simulated taking into account quadrupole interactions, is shown by the black line. Grey and black arrows indicate quadrupole satellite and homonuclear absorption peaks, respectively.

warm-up spectrum in a zero magnetic field is best described by a model in which all spin clusters are taken into account simultaneously. Consideration of NSS in the form of individual nuclear spin clusters is possible, first of all, due to the low abundance of magnetic isotopes in CdTe. Each cluster contains one Te^{125} nucleus and from one to four Cd^{111}/Cd^{113} nuclei, which are the closest neighbors to the Te^{125} atom. The simulated warm-up spectrum in a zero magnetic field as part of the nuclear spin cluster model, shown in Figure 1, *b* by the black line, describes the experimental spectrum for the CdTe QW (green dots) quite well.

It is noteworthy that the proposed cluster model also describes well the warm-up spectra measured in external static magnetic fields. From the work by A. Nolle [14] it is known that in NMR spectra the main absorption peaks corresponding to the precession of the spins of the Cd and Te isotopes are accompanied by spin-spin satellites. In the warm-up spectra we obtained in external fields, there are also these satellites, and their frequency positions are quite accurately described by the proposed cluster model. An example of the warm-up spectrum we measured in an external field $B_x = 14.5$ G and its theoretical simulation are shown in Figure 2, *a* with blue dots and a black line, respectively. Grey arrows indicate the positions of spin-spin satellites, black arrows indicate the positions of the main homonuclear absorption peaks. A detailed description of the model, as well as a comparison of the simulated and experimental spectra in a wide range of external static magnetic fields for a CdTe heterostructure are presented in the work [18].

For visual comparison Figure 2, *b* shows the measured warm-up spectrum in a static magnetic field $B_x = 14.5$ G for a deformed bulk layer *n*-GaAs (blue dots) [16]. An external field of this magnitude is not yet sufficient to separate one observed broad absorption peak into individual homonuclear peaks corresponding to the precession of the isotopes As^{75}, Ga^{69} and Ga^{71}, as well as into quadrupole satellites. An example of a warm-up spectrum measured in an external static magnetic field $B_x = 48.3$ G for a deformed bulk layer *n*-GaAs, in which individual peaks are visible, is shown in Figure 3 with blue dots. The black line shows the simulated spectrum. Figure 3 demonstrates broad, compared to the absorption peaks for CdTe QWs (HWHM ~ 1.5 kHz), homonuclear peaks (black arrows in Figure 3), accompanied by quadrupole satellites, which are caused by mechanical stress in the structure (their positions are indicated by grey arrows in Figure 3).

4. Conclusion

This work presents a review of the main experimental and theoretical results that we obtained earlier, which describe the manifestation of nuclear spin-spin interactions in the warm-up spectra in zero and weak external magnetic fields. A distinctive feature of our experiments is the ability to measure the warm-up spectrum of a cooled NSS in a zero magnetic field, when the nuclei do not participate in the Zeeman interaction and the spectrum is determined by purely nuclear spin-spin interactions. Spectra in weak external static magnetic fields up to 50 G are also presented and analyzed.

The results are presented using the example of two semiconductor structures: a CdTe(CdZnTe) heterostructure and a deformed bulk layer of *n*-GaAs. The choice of these materials is primarily due to their different nuclear spin properties, which, as shown in this review, are clearly manifested in the warm-up spectra. This is primarily due to the weak isotopic abundance in CdTe, which, in particular, allowed us to use the nuclear spin cluster model to describe the measured spectra. Meanwhile, the shape of the warm-up spectra for the deformed bulk layer *n*-GaAs is mainly determined by quadrupole effects caused by mechanical stresses in the crystal.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] Optical Orientation, edited by F. Meier and B.P. Zakharchenya (North Holland, Amsterdam, 1984).
[2] M. Goldman. Spin Temperature and Nuclear Magnetic Resonance in Solids. Oxford University Press (1970).

[3] V.K. Kalevich, V.D. Kulkov, V.G. Fleisher. Pis'ma v ZHETF, **35**, 17 (1982). (in Russian).
[4] V.K. Kalevich, V.G. Fleischer. Izv. AN SSSR. Ser. fiz. **47**, *12*, 2294 (1983). (in Russian).
[5] I.A. Merkulov. FTT **40**, *6*, 1018 (1998). (in Russian).
[6] D. Scalbert. Phys. Rev. B **95**, 245209 (2017).
[7] M. Vladimirova, D. Scalbert, M.S. Kuznetsova, K.V. Kavokin. Phys. Rev. B **103**, 205207 (2017).
[8] V.M. Litvyak, R.V. Cherbunin, V.K. Kalevich, K.V. Kavokin. Phys. Rev. B **108**, 235204 (2023).
[9] V.M. Litvyak, R.V. Cherbunin, K.V. Kavokin, V.K. Kalevich. IOP Conf. Ser.: J. Phys. **951**, 012006 (2018).
[10] M. Kotur, D.O. Tolmachev, V.M. Litvyak, K.V. Kavokin, D. Suter, D.R. Yakovlev, M. Bayer. Commun. Phys. **4**, *1*, 193 (2021).
[11] R.G. Shulman, J.M. Mays, D.W. McCall. Phys. Rev. **100**, 692 (1955).
[12] R.G. Shulman, B.J. Wyluda, H.J. Hrostowski. Phys. Rev. **109**, 808 (1958).
[13] R.K. Hester, A. Sher, J.F. Soest, G. Weisz. Phys. Rev. B **10**, 4262 (1974).
[14] A. Nolle. Z. für Physik B Condens. Matter **34**, 175 (1979).
[15] M.M. Sobolev, P.N. Brunkov, S.G. Konnikov, M.N. Stepanova, V.G. Nikitin, V.P. Ulin, A.S. Dolbaya, T.D. Kamushadze, R.M. Masuradze. Sov. Phys. Semicond. **23**, 660 (1989).
[16] V.M. Litvyak, R.V. Cherbunin, V.K. Kalevich, A.I. Lihachev, A.V. Nashchekin, M. Vladimirova, K.V. Kavokin. Phys. Rev. B **104**, 235201 (2021).
[17] V.M. Litvyak, R.V. Cherbunin, V.K. Kalevich, K.V. Kavokin. Semiconductors **54**, 1728 (2020).
[18] V.M. Litvyak, P. Bazhin, R. André, M. Vladimirova, K.V. Kavokin. arXiv:2403.17593 [cond-mat.mes-hall] (2024).
[19] B.F. Gribakin, V.M. Litvyak, M. Kotur, R. Andre, Ḿ. Vladimirova, D.R. Yakovlev, K.V. Kavokin. Phys. Rev. B **109**, (2024).
[20] V.M. Litvyak, R.V. Cherbunin, F.Yu. Soldatenkov, V.K. Kalevich, K.V. Kavokin. St. Petersburg Polytechnic University J. Physics and Mathematics **16** (2023).
[21] A.V. Mikhailov, A.S. Kurdyubov, E.S. Khramtsov, I.V. Ignatiev, B.F. Gribakin, S. Cronenberger, D. Scalbert, M.R. Vladimirova, R. Andre arXiv:2304.07135 (2023).
[22] Y.M. d'Aubigne, H. Mariette, N. Magnea, H. Túffigo, R.T. Cox, G. Lentz, L.S. Dang, J.L. Pautrat, A. Wasiela. Surface Science **101**, 650 (1990).

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