

Measurement of nuclear local field by adiabatic demagnetization method for CdTe/CdZnTe quantum well

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This paper is devoted to measurement of the nuclear local field in a CdTe/CdZnTe quantum well. The features of this structure are a weak natural abundance of magnetic isotopes, as well as the absence of quadrupole moments of nuclear spins. These characteristics of the nuclear spin system affect the magnitude of the nuclear local field, as well as the methods of its measurement under optical experimental conditions. We used method based on adiabatic demagnetization of an optically cooled system of nuclei. A comparative analysis of the local field measured in this work with previous results on measuring the absorption spectrum of an oscillating magnetic field power was also carried out. The local field estimate from the spectrum line width gave an underestimated value compared to that obtained in adiabatic demagnetization experiments. A theoretical calculation of the local field was performed, which also took into account all possible nuclear spin-spin interactions for CdTe with known values of the constants. The calculation results are consistent with the experimental data.

Keywords: CdTe/CdZnTe quantum well, nuclear local field, nuclear spin-spin interactions.

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1. Introduction

In solids at low temperatures of the crystal lattice, nuclear spins form an energetically isolated system obeying the laws of thermodynamics [1]. The most important parameter of a nuclear spin system, which determines its thermodynamic properties in weak external magnetic fields, is the local field [2], which characterizes nuclear spin-spin interactions. These include magnetodipole interactions, as well as indirect interactions through valence electrons (exchange and pseudodipole). The magnetic dipole-dipole interaction of the magnetic moments of nuclei is the most long-range type of nuclear spin-spin interactions [1,3]. There are also indirect internuclear interactions through valence electrons, which are short-range, i.e., nonzero only for the nearest neighbors. Nuclear spin-spin interactions in semiconductor crystals of groups III–V and II–VI have been experimentally studied by broadening absorption lines in nuclear magnetic resonance (NMR) spectra in strong external magnetic fields (significantly exceeding nuclear local fields).

For CdTe, one of the first experimental manifestations of magnetodipole and indirect nuclear spin interactions was presented in the study of A. Nolle [4]. The author conducted NMR experiments at resonant frequencies of ^{113}Cd and ^{125}Te isotopes for different orientations of the external magnetic field. It has been shown that the splitting of resonant absorption lines of ^{113}Cd and ^{125}Te isotopes occurs due to dipole-dipole, as well as indirect interactions between ^{125}Te and $^{111,113}\text{Cd}$.

It is also known from the work of [5] that the direct dipole-dipole coupling in CdTe is weak due to the low

natural content of magnetic isotopes, which include $^{123,125}\text{Te}$ and $^{111,113}\text{Cd}$. This leads, in particular, to narrow absorption lines, from the half-widths of which it is possible to estimate the dipole contribution to the magnitude of the nuclear local field.

For different semiconductors, the constants of indirect interactions can differ both in magnitude and in sign. This can lead to suppression or enhancement of the main magnetodipole interaction, which primarily affects the magnitude of the local field. The values of the constants of nuclear spin-spin interactions for CdTe were used to calculate the nuclear local field and are given in Ref. [6].

In addition to spin-spin interactions, when estimating the values of local fields, possible quadrupole interactions should be taken into account. If the spin value of the nuclear isotope is $I > 1/2$, then these isotopes can participate in quadrupole effects. The presence of quadrupole interactions in the semiconductor structure leads to an increase in the purely spin-spin local field:

$$B_L = \sqrt{B_{SS}^2 + B_Q^2}, \quad (1)$$

where B_{SS} and B_Q is the spin-spin and quadrupole contributions to the magnitude of the local field B_L .

Quadrupole interactions are particularly pronounced in stressed semiconductor structures such as quantum wells, quantum dots, and semiconductor structures embedded in microresonators. Pronounced uniaxial deformation occurs in these heterostructures due to differences in the permanent lattices of the semiconductors from which the sample is grown. This deformation leads to the formation of an electric field gradient (EFG), with which nuclear isotopes

with spin values $I > 1/2$ interact. This leads to an increase in the local field, which was experimentally shown in Refs. [7,8].

In the CdTe/CdZnTe quantum well (QW) studied in this paper, all magnetic isotopes ($^{111,113}\text{Cd}$ and $^{123,125}\text{Te}$) have spin $I = 1/2$ and do not participate in quadrupole interactions. Therefore, the local field is formed only by spin-spin interactions: $B_L = B_{SS}$.

To date, there are practically no experimental and theoretical studies aimed at directly determining such a fundamental characteristic of the nuclear spin system (NSS) in CdTe as the nuclear local field. However, it is very important to be able to determine its value for the studied crystal, since B_L forms the behavior of the NSS in zero ($B_{\text{ext}} = 0$) and weak external magnetic fields ($B_{\text{ext}} < B_L$). In particular, the local field determines the heat capacity and susceptibility of the NSS, as well as the minimum limit of the nuclear spin temperature Θ_f in experiments on adiabatic demagnetization [1,3]:

$$\Theta_f = \Theta_i \frac{B_L}{B_Z}, \quad (2)$$

where Θ_i is the value of the nuclear spin temperature obtained after optical cooling in the longitudinal optical pumping field B_Z .

This paper presents the results of measurements of the magnitude of the nuclear local field using an experimental technique involving adiabatic demagnetization of an optically cooled liquid. This technique makes it possible to obtain the magnitude of the nuclear local field, taking into account all possible spin interactions of the nuclei. The measured value of the local field is consistent with the theoretical calculation. It will also be shown that for a semiconductor structure with a low abundance of magnetic isotopes and in the absence of nuclear quadrupole interactions, the magnitude of the local field estimated from the width of the heating spectrum lines turns out to be less than that measured using adiabatic demagnetization. This is due to the fact that the width of the absorption line is formed only by the long-range dipole coupling of nuclear spins in CdTe, and indirect interactions, which are the main ones in CdTe, do not contribute to the broadening. It should be noted that the results of experiments on adiabatic demagnetization, which include measurements of both the local field and the warming spectra, are ways to estimate the values of nuclear spin interactions in the studied structure.

2. Sample and experimental setup

In most semiconductor structures, in particular in cadmium telluride, the rates of spin-spin and spin-lattice relaxation of nuclei can differ by several orders of magnitude, which makes it possible to lower the NSS temperature to several microkelvins. In this case, the studied sample is at temperatures close to the temperature of liquid helium. The temperature of NSS in semiconductor structures 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 [1,3]. 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. This feature of the NSS is the basis of the methodology used in our work to measure the nuclear local field. It should be noted that this technique has also been used in previous studies on measuring the absorption spectra of the alternating magnetic field of an optically cooled NSS [6,9]. The previously measured spectrum in the zero static magnetic field [6] will be considered in this paper in order to estimate the magnitude of the local field from its width and compare it with the results of adiabatic demagnetization.

The studied structure is a single CdTe/CdZn_{0.05}Te_{0.95} 30 nm wide cell grown by molecular beam epitaxy on a substrate CdZn_{0.04}Te_{0.96} (100). The electron-nuclear spin dynamics in this sample was studied in detail in Refs. [6,10].

The studied sample was placed in a closed-cycle cryostat and cooled to a temperature of 10 K. Radiation from a laser diode at a wavelength of 680 nm passed through a linear polarizer and a quarter-wave plate ($\lambda/4$), creating a circularly polarized optical pump, and then focused on the sample surface. The polarized photoluminescence (PL) signal from the sample was formed by a lens into a parallel beam of light, which passed through a photoelastic modulator (PEM), a linear polarizer and focused on the slit of the spectrometer. The spectrometer transmitted PL intensity at a wavelength of 774 nm, which was then recorded using an avalanche photodiode and a two-channel photon counter synchronized with PEM.

3. Methodology for measuring the local field in experiments on adiabatic demagnetization

The local field measurements are based on the adiabatic demagnetization method, which makes it possible to directly obtain the magnitude of the local field from experiments. With an adiabatic (i.e., slow on the time scale of spin-spin relaxation, but fast on the scale of spin-lattice relaxation of nuclei) change in the external magnetic field, the nuclear spin polarization changes according to the law [2]:

$$\frac{P_N}{P_{Ni}} = \frac{B}{B_i} \sqrt{\frac{B_L^2 + B_i^2}{B_L^2 + B^2}}, \quad (3)$$

where P_N and P_{Ni} is the current and initial values of the nuclear spin polarization, B and B_i is the current and initial values of the external magnetic field.

Detailed details of the experimental technique for adiabatic demagnetization of optically cooled plastics and processing of experimental results in order to determine the magnitude of the local field are given in Ref. [11] for a bulk sample *n*-GaAs. This article briefly discusses the main features of this technique when used for CdTe.

To measure local fields, a three-stage optical protocol was implemented based on measuring the degree of circular polarization of PL. At the first stage, optical cooling of the NSS was carried out by circularly polarized pumping in a longitudinal magnetic field $B_Z = 150$ Gc for $t_{\text{pump}} = 500$ s. Optical cooling was followed by adiabatic demagnetization in a local field: the longitudinal field B_Z was turned off to zero for 100 ms. At the third stage, the transverse field B_x was turned on, in which the nuclear field B_N was measured. About ten to fifteen three-stage measurements were made for various values B_x (from 0.2 to 10 Gs). As a result of the analysis of experimental data, the dependence $B_N(B_x)$ was constructed.

An example of the time dependence of the degree of polarization of PL $\rho(t)$, measured according to the three-stage protocol, is shown in Figure 1 with blue dots for the measuring field $B_x = 1$ Hs. The red curve in Figure 1 shows an approximation of the dependence $\rho(t)$ according to the formula (4) with the value of the nuclear field B_N as a free parameter:

$$\rho(t) = \rho_0 \frac{B_{1/2}^2}{B_{1/2}^2 + (B_x + b + (B_N - b) \exp(-t/T_1))^2}, \quad (4)$$

where $\rho_0 = 6.7\%$ is the magnitude of the degree of polarization of PL at the optical cooling stage, $B_{1/2} = 40$ Gc is the half-width at the half-height of the PL depolarization curve in a transverse magnetic field with rapid modulation of the pump polarization (Hanle curve), $T_1 = 30$ s is the time of spin-lattice relaxation of nuclei, b is the nuclear field resulting from dynamic nuclear polarization at the measurement stage. The values $B_{1/2}$ and T_1 were determined in separate experiments.

It should be noted that, unlike the method used in Ref. [12], the adiabatic demagnetization method does not require the determination of the factor ξ and directly gives the magnitude of the local field.

4. Experimental results

4.1. Determination of the magnitude of the local field from an experiment on adiabatic demagnetization

As a result of processing three-stage time dependences of the degree of polarization of PL for various values of measuring fields B_x , the dependence of the values of nuclear fields on the field B_x , $B_N(B_x)$ was obtained. The obtained dependence for CdTe/CdZnTe QE is shown by dots in Figure 2. The line corresponds to the approximation of

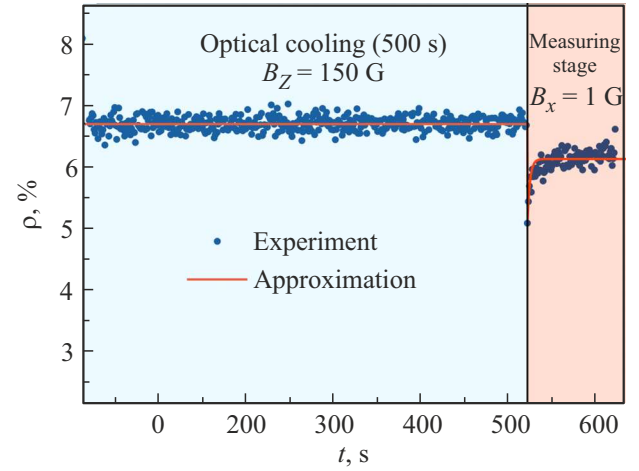


Figure 1. Time dependence of the degree of polarization of PL for CdTe/CdZnTe (blue dots). The optical cooling stage is highlighted by blue color, and the measurement stage is highlighted by pink color. The intermediate stage of adiabatic demagnetization is not visible on the curve due to the short duration (100 ms). The obtained dependence is approximated by the formula (4) (red line) with the magnitude of the nuclear field as the free parameter.

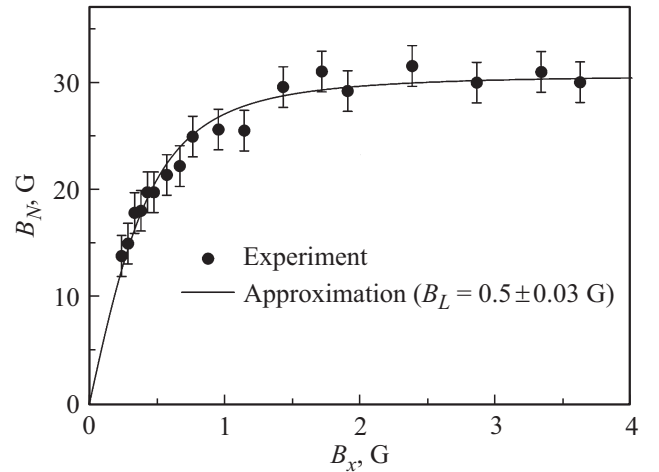


Figure 2. The experimentally measured dependence of the nuclear field B_N on the magnitude of the measuring transverse magnetic field B_x for CdTe/CdZnTe is represented by dots. Line — approximation by the formula (3) with the value of the local field $B_L = 0.5 \pm 0.03$ Gs.

the experimental points according to the formula (3) with the value of the local field $B_L = 0.5 \pm 0.03$ Gs.

4.2. Estimation of the magnitude of the local field from the width of the absorption lines

The papers in Refs. [6,9] are devoted to the measurement of the absorption spectra of the alternating magnetic field power of an optically cooled NSS, as well as theoretical interpretations of the frequency positions of absorption peaks for a bulk *n*-GaAs sample and for CdTe/CdZnTe QW.

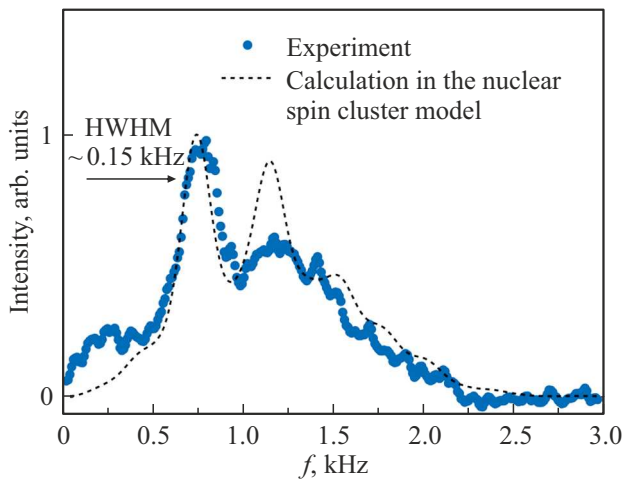


Figure 3. The warming spectrum upon absorption of AC magnetic field power, measured in a zero static magnetic field for CdTe/CdZnTe QE (points). The values of the heating rates in relative units are plotted along the ordinate axis. The dotted line shows the spectrum [6] calculated within the framework of the nuclear spin cluster model.

However, the studies conducted in these papers did not aim to estimate the magnitude of the local field based on the shapes of the observed absorption peaks. It is known from NMR theory that the width of absorption lines is related to internuclear spin interactions, which include both pure spin-spin and quadrupole [13]. Previously measured spectra of heating in a zero magnetic field for the bulk structure *n*-GaAs in the framework of the NSS [9] heating spectroscopy had a half-width at half-height HWHM (half width at half maximum) ~ 2 kHz. At the same time, for the deformed sample, quadrupole interactions were manifested in the heating spectrum as two absorption peaks with HWHM ~ 2 kHz at isotope precession frequencies ^{75}As and $^{69,71}\text{Ga}$ in local fields determined by spin-spin and quadrupole interactions (Figure 1 in Ref. [14]). For a sample with a minimum amount of residual deformation, the heating spectrum contains one peak.

We also made a series of measurements of the heating spectra both in zero (Figure 3) and in external magnetic fields [6] in order to obtain spectra taking into account only the purely spin-spin interaction, when quadrupole effects are precisely absent. The method of measuring the heating spectra is also based on the preliminary optical cooling of the NSS followed by adiabatic demagnetization. But an additional stage of exposure to an alternating magnetic field of a given power was added before the measuring stage. An unexpectedly complex view of the heating spectra was obtained, which was explained by us in the framework of the nuclear spin cluster model [6]. But in this paper, we only care about the half-width of the absorption line, from which an estimate of the local field will be obtained. Therefore, we will omit the details of the analysis of experimental data to obtain the warming spectra.

The HWHM value was ~ 0.15 kHz from the approximation of the absorption lines of the heating spectrum measured in a zero magnetic field for CdTe/CdZnTe QW. Then, according to the estimation formula $B_L \sim \text{HWHM}/(\langle\gamma_N\rangle) = 0.2$ G, where $\langle\gamma_N\rangle$ is the average gyromagnetic ratio of magnetic isotopes.

Thus, from the estimate of the width of the absorption lines, the local field was ~ 2.5 times less than from direct measurements by adiabatic demagnetization. We explain this difference by the fact that demagnetization allows obtaining a local field, which is formed by both dipole-dipole and indirect nuclear spin interactions, which give a value of $B_L = 0.5 \pm 0.03$ G. The experimental result obtained is confirmed by a theoretical calculation, the details of which are given in sec. 5 of this article.

The model of nuclear spin clusters developed by us in Ref. [6] explains the fact that the widths of the observed peaks in the thaw spectrum are formed only by long-range dipole coupling (random nuclear fields from single nuclear spins not included in clusters). The fine structure of the spectrum is formed by both indirect and dipole interactions within nuclear spin clusters. Therefore, the estimate of the absorption line width gives an incomplete value of the nuclear local field.

5. Theoretical calculation of the magnitude of the nuclear local field for CdTe

As noted above, the measured value of the nuclear local field in experiments on adiabatic demagnetization is formed by all types of nuclear spin interactions characteristic of the studied structure. For CdTe, due to the low prevalence of magnetic isotopes, indirect interactions of nuclear spins occurring through valence electrons dominate. The dipole-dipole interaction is weak in CdTe because it is long-range. However, it cannot be excluded from consideration. Based on this, a theoretical calculation of the local field of nuclear spin-spin interactions was carried out, taking into account both the dipole-dipole and the dominant indirect (pseudodipole and exchange) nuclear spin interactions. The calculation used constants for CdTe isotopes taken from Ref. [4].

The local field is defined as the square root of the ratio of the square of the average energy of nuclear spin-spin interactions, with the exception of the Zeeman interaction, and the average square of one of the components of the magnetic moment:

$$B_L^2 = \frac{\text{Tr}(H_{ss}^2)}{\text{Tr}(M_z^2)} = \frac{3}{\text{Tr}(1)N(I+1)\sum_{\alpha} x_{\alpha}(\hbar\gamma_{\alpha})^2} \text{Tr}(H_{ss}^2). \quad (5)$$

Here I is the spin of the nuclei (in CdTe, the spins of all magnetic nuclei are the same and equal to $1/2$), γ_{α} is gyromagnetic isotope ratio α , x_{α} is proportion of nuclei of α isotope in the sample, N is the total number of nuclei [2].

Values of the relative number of nuclei, gyromagnetic constants of each isotope, and constant indirect interactions

	¹¹³ Cd	¹¹¹ Cd	¹²⁵ Te	¹²³ Te
x_α , %	6.1	6.4	3.0	0.45
γ_α , rad · kHz/G	−5.97	−5.72	−8.48	−7.04
$A(^{125/123}\text{Te}_\alpha)/h$, kHz	0.655 [4]	0.622 [4]	—	—
$B(^{125/123}\text{Te}_\alpha)/h$, kHz	0.110 [4]	0.101 [4]	—	—

Spin-spin interactions between magnetic nuclei in CdTe can be divided into two groups: direct dipole-dipole and indirect. The direct dipole-dipole interaction between a pair of nuclei is given by the formula

$$H_{1-2}^{dd} = \frac{\hbar^2 \gamma_1 \gamma_2}{r_{12}^3} \left[\left((\vec{I}_1 \vec{I}_2) - 3(\vec{R}_{12} \vec{I}_1)(\vec{R}_{12} \vec{I}_2) \right) \right], \quad (6)$$

where r_{12} is the distance between the cores, $\vec{R}_{12} = \vec{r}_{12} / r_{12}$.

Indirect interactions take place through valence electrons, so they occur only between the nearest neighbors. By symmetry, they are divided into scalar (exchange) and pseudodipole indirect interactions. Their Hamiltonians have the form:

$$H_{1-2}^{sc} = A_{12} \delta_{r_{12}, r_0} (\vec{I}_1 \vec{I}_2),$$

$$H_{1-2}^{pdd} = B_{12} \delta_{r_{12}, r_0} \left[\left((\vec{I}_1 \vec{I}_2) - 3(\vec{R}_{12} \vec{I}_1)(\vec{R}_{12} \vec{I}_2) \right) \right], \quad (7)$$

where A_{12} and B_{12} are the constants of scalar and bipolar indirect interactions, respectively, $r_0 = \frac{\sqrt{3}}{4} a$ are the distance between the nearest neighbors.

Then the complete spin-spin interaction can be described by the following:

$$H_{ss} = \sum_\alpha \sum_{i < j} \frac{\hbar^2 \gamma_\alpha^2}{r_{ij}^3} \left[(\vec{I}_i^\alpha \vec{I}_j^\alpha) - 3(\vec{R}_{ij} \vec{I}_i^\alpha)(\vec{R}_{ij} \vec{I}_j^\alpha) \right]$$

$$+ \sum_\alpha \sum_{b > \alpha} \sum_i \sum_j \left\{ \left(\frac{\hbar^2 \gamma_\alpha \gamma_\beta}{r_{ij}^3} + B_{\alpha\beta} \delta_{r_{ij}, r_0} \right) \right.$$

$$\times \left[(\vec{I}_i^\alpha \vec{I}_j^\beta) - 3(\vec{R}_{ij} \vec{I}_i^\alpha)(\vec{R}_{ij} \vec{I}_j^\beta) \right] + A_{\alpha\beta} \delta_{r_{ij}, r_0} (\vec{I}_i^\alpha \vec{I}_j^\alpha) \left. \right\}. \quad (8)$$

Here, the magnetic isotopes are indicated by the Greek indexes, and the nuclei of the corresponding isotopes are indicated by the Latin indexes. Raising (8) to the square and tracking it using the ratio

$$\text{Tr}((I_\alpha^i)_s (I_\beta^j)_p) = \delta_{\alpha, \beta} \delta_{i, j} \delta_{s, p} \frac{I(I+1)(2I+1)}{3},$$

we obtain

$$\text{Tr}(H_{ss}^2) = \frac{NI^2(I+1)^2}{3} \text{Tr}(1) \sum_\alpha x_\alpha \left\{ \hbar^4 \gamma_\alpha^4 \sum_{i < j} \frac{1}{r_{0j}^6} \right.$$

$$+ \sum_{\beta > \alpha} \sum_j \left[2 \left(\frac{\hbar^2 \gamma_\alpha}{r_{0j}^3} + B_{\alpha\beta} \delta_{r_{0j}, r_0} \right)^2 + (A_{\alpha\beta} \delta_{r_{0j}, r_0})^2 \right] \left. \right\}. \quad (9)$$

Then the expression for the square of the local field has the form

$$B_L^2 = \frac{I(I+1)}{\sum_\alpha x_\alpha \hbar^2 \gamma_\alpha^2} \sum_\alpha x_\alpha \left\{ \hbar^4 \gamma_\alpha^4 \sum_{i < j} \frac{1}{r_{0j}^6} \right.$$

$$+ \sum_{\beta > \alpha} \sum_j \left[2 \left(\frac{\hbar^2 \gamma_\alpha}{r_{0j}^3} + B_{\alpha\beta} \delta_{r_{0j}, r_0} \right)^2 + (A_{\alpha\beta} \delta_{r_{0j}, r_0})^2 \right] \left. \right\}. \quad (10)$$

The values of the gyromagnetic ratios γ_α , the constant indirect interactions A and B , and the relative number of nuclei x_α for each isotope of the variety α are presented in the table.

The calculation gives the value of the local field for CdTe: $B_L = 0.4 \pm 0.02$ G. This local field value is close to that obtained in the adiabatic demagnetization experiment.

6. Conclusion

Experimental results of measuring the nuclear local field during adiabatic demagnetization of optically cooled YASS in a CdTe/CdZnTe quantum well are presented in this paper. This experimental technique makes it possible to determine the magnitude of the nuclear local field, taking into account the contributions from all the nuclear spin interactions available for this structure. None of the magnetic isotopes in CdTe has a quadrupole moment, so the local field is created only by nuclear spin-spin interactions. Also, due to the weak natural abundance of magnetic isotopes in CdTe, the local field is formed primarily by short-range indirect interactions of nuclear spins through valence electrons (pseudodipole and exchange), as well as long-range magnetodipole interactions. The measurements determined the local field strength of ~ 0.5 G. The theoretical calculation, taking into account all the nuclear spin-spin interactions present in CdTe, gave a local field value of 0.4 G, which is in good agreement with the experimental result. It has also been found that the long-range dipole interaction contributes to the local field of ~ 0.2 G, and its magnitude can be estimated from the width of the AC magnetic field power absorption line measured in a zero static field.

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Conflict of interest

The authors declare that they have no conflict of interest.

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