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We show that the spin-lattice relaxation in *n*-type insulating GaAs is dramatically accelerated at low magnetic fields. The origin of this effect, which cannot be explained in terms of well-known diffusion-limited hyperfine relaxation, is found in the quadrupole relaxation, induced by fluctuating donor charges. Therefore, quadrupole relaxation, which governs low field nuclear spin relaxation in semiconductor quantum dots, but was so far supposed to be harmless to bulk nuclei spins in the absence of optical pumping, can be studied and harnessed in the much simpler model environment of *n*-GaAs bulk crystal.

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Introduction. Understanding and manipulating nuclear magnetization in the vicinity of semiconductor-hosted defects is an issue of both technological and fundamental importance [1–4]. It concerns many different systems, such as semiconductor quantum dots [2], nitrogen-vacancy centers in diamond [5], silicon-vacancy centers in SiC [6], and other systems, where an electron spin can be used to transfer angular momentum from light onto nuclei, and nuclei can store information in their spin degree of freedom. This is possible because a nuclear spin system (NSS) is weakly coupled to the lattice [7–9]. The fact that equilibrium within the NSS is established much faster than the equilibrium with the crystal lattice justifies its thermodynamic description and the concept of nuclear spin temperature Θ_N [10]. Early studies on bulk semiconductors demonstrated that combining optical pumping under magnetic fields above local field B_L (given by magnetic interactions within the NSS) with nuclear adiabatic demagnetization to the fields $B < B_L$, it is possible to cool the NSS well below lattice temperature [8,9,11]. Various thermodynamic transitions to spin-ordered states were theoretically predicted at $\Theta_N < 1\ \mu$K [12,13]. Such degree of control over NSS would open new possibilities for semiconductor spintronics, where fluctuations in the nuclear spin system are considered as a major and ubiquitous source of decoherence [14,15].

However, deep cooling of the NSS is challenging, limited by two main issues: the efficiency of the NSS pumping and the relaxation during the demagnetization stage. It was suggested that light-induced nuclear quadrupolar relaxation can strongly reduce the pumping efficiency [16]. Even so, the relaxation of NSS in the absence of optical pumping in a range of magnetic fields down to zero field (i.e., at the conditions met during the demagnetization) has not been addressed so far. Two different regimes of relaxation should be considered [Fig. 1(a)]. At strong magnetic fields the projection of the nuclear angular momentum onto the field is a conserved quantity. In this regime the energy of the NSS is determined by the Zeeman interaction, so that spin-lattice relaxation involves changing of both energy and angular momentum, mediated by non-spin-conserving interactions. At low fields, $B \leqslant B_L$, the nonequilibrium nuclear angular momentum decays within the spin-spin relaxation time T_2 of order of 100 μs.

In this regime the polarization of the NSS is induced by the external field via its paramagnetic susceptibility. The susceptibility χ is inversely proportional to Θ_N, which relaxes towards the lattice temperature T with the characteristic spin-lattice relaxation time $T_1 \gg T_2$.

Therefore, the nuclear spin relaxation at low field is in fact the warm up of the NSS, which is determined by energy transfer between the NSS and the crystal lattice.

This warm up may present a fundamental obstacle on the way towards ultralow temperatures in NSS.

In this Rapid Communication we investigate the NSS warm up in bulk *n*-GaAs using photoluminescence (PL) spectroscopy at various magnetic fields and temperatures. We show that the change of the nuclear spin-lattice relaxation regime from angular momentum relaxation at $B > B_L$ to the NSS warm up at $B < B_L$ is accompanied by a dramatic increase of the relaxation rate $1/T_1$, Fig. 1(a). This behavior is completely unexpected [17] within the standard model of the diffusion-limited hyperfine-induced NSS relaxation [18–20]. We suggest that low-field relaxation is due to the interaction of nuclear quadrupole moments with electric field gradients induced by slow spatiotemporal fluctuations of localized electron charges, Fig. 1(b). These fluctuations are characterized by the electron charge correlation time $\tau_c^c \gg T_2$, and result from the electron hopping either into conduction band, or between the donor sites, as evidenced by the measurements of resistance as a function of temperature. Our theory shows that the energy flux between nuclear spin and electron charge via slowly varying quadrupole interaction $\mathcal{F}_Q$ does not depend on the magnetic field, while NSS heat capacity is strongly field dependent [21]. This explains the strong field dependence of the NSS warm-up rate. The model provides a quantitative understanding of the experimental results and suggests the pathways for the optimization of the NSS cooling.

Sample and experimental setup. We have studied a 20-μm-thick GaAs sample grown by liquid-phase epitaxy, with a Si donor concentration of $n_d = 4\times10^{15}$ cm^3, already used in a previous work [23]. The sample was placed in a variable temperature or He bath cryostat, surrounded by an electromagnet. The magnetic field was applied in the oblique but nearly Voigt geometry ($<10^\circ$), in order to allow for both dynamic nuclear polarization and detection due to Hanle effect induced by nuclear spin. The sample was excited by a Ti-sapphire laser beam tuned at $E = 1.55$ eV, circularly polarized, and focused on a 50-μm-diameter spot. The PL

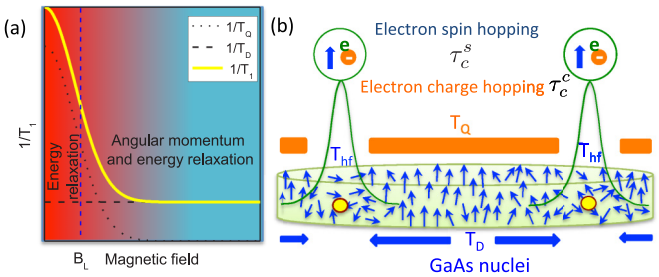

FIG. 1. Sketch of the NSS warm up. (a) Magnetic field dependence of the two contributions to the nuclear spin-lattice relaxation: diffusion-limited hyperfine interaction at rate $1/T_D$ (dashed line) and quadrupole-induced warm up at rate $1/T_Q$. (b) Relevant processes in a nuclei-electron coupled system: fast nuclear spin warm up under donor orbits via hyperfine interaction (characterized by time T_{hf}), and warm up of all other nuclei via spin diffusion towards the donor sites and quadrupole interaction.

was collected in the reflection geometry, passed through a circular polarization analyzer [consisting of a photoelastic modulator (PEM) and a linear polarizer], spectrally dispersed with a double-granting spectrometer, and detected by an avalanche photodiode, connected to a two-channel photon counter synchronized with the PEM.

Experimental results. The three-stage measurement protocol is illustrated in Fig. 2. During the pumping stage, both magnetic field $B_{\text{pump}} = 3.5$ G and the pumping beam are switched on, providing the cooling of the NSS. The duration of this stage is fixed to $T_{\text{pump}} = 5$ min. After T_{pump}, the pump beam is switched off, and the magnetic field is set to the value B_{dark} at which we want to study the warm up of the NSS. The second stage of the experiment will be referred to as a dark stage; its duration T_{dark} was varied in order to access the corresponding variation of the nuclear spin polarization. Immediately [on the scale of electron spin relaxation time ($T_s \approx 100$ ns [23]] after switching off the pump, electron spin polarization returns to its equilibrium state, determined by the sum of the applied magnetic field and the Overhauser field created by the NSS. The NSS warm up is much slower, $T_1 \gg T_s$. In order to measure the evolution of NSS polarization in the dark, Overhauser field $B_N(T_{\text{dark}})$ achieved after T_{dark} is measured during the third stage of the protocol. The light is switched back on, measuring field B_{pump} is restored, and the circular polarization degree of the PL is detected as a function of time during 150 s. The value of the PL polarization degree ρ_{dark} in the beginning of the third stage is extracted from the exponential fit of this decay, for each value of T_{dark} [Fig. 2(c)]. The corresponding value of B_N, which is proportional to the inverse nuclear spin temperature $\beta = 1/k_B\Theta_N$, is related to ρ_{dark} via Hanle formula

$$\beta(T_{\text{dark}}) \propto B_N(T_{\text{dark}}) = B_{1/2}\sqrt{\frac{\rho_0 - \rho_{\text{dark}}}{\rho_{\text{dark}}}} - B_{\text{pump}}, \qquad (1)$$

where ρ_0 is the PL polarization in the absence of the external field, and $B_{1/2}$ is the half width of the Hanle curve, measured independently [24] under conditions where nuclear spin polarization is absent (pump polarization is modulated at 50 kHz). Thus, measuring ρ_{dark} for different waiting times T_{dark} we can follow nuclear spin warmnup or, equivalently, the evolution of the Overhauser field B_N, Fig. 2(d). Fitting the result to the exponential decay (growth for Θ_N), we obtain the NSS relaxation time T_1 for a given external magnetic field B_{dark} [Fig. 2(d)] [25]. A similar protocol has been first proposed and realized by Kalevich *et al.* [11], and then further developed in Ref. [24].

Figure 3(a) summarizes the magnetic field dependence of the NSS warm-up rate T_1 measured at different temperatures. Solid lines are fits to the Lorentz shape with the half width at half maximum (HWHM) reported as color-encoded symbols in Fig. 3(b), while the NSS warm-up times at $B = 0$ and $B = \infty$ are reported in Fig. 3(c). The salient feature of these data

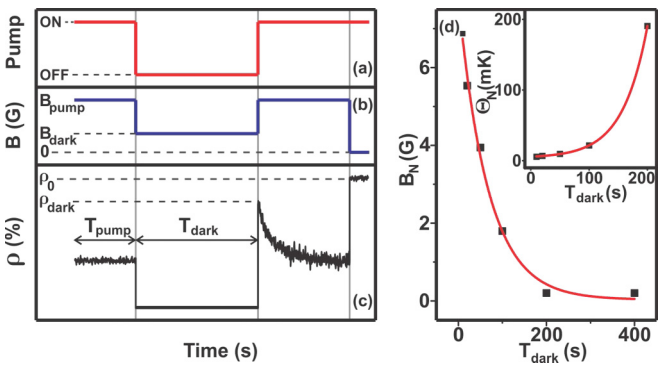

FIG. 2. A scheme of the three-stage experimental protocol. Each stage is characterized by (a) the presence/absence of the pump, and (b) the value of the magnetic field applied during the dark stage. (c) The corresponding values of the PL polarization degree. (d) A set of measurements of $\rho_{\text{dark}} \propto B_N$ for a given applied field $B_{\text{dark}} = 8$ G, obtained at different values of dark intervals T_{dark} (symbols). The solid line is the fit to the exponential decay, which determines the NSS warm-up time T_1. Inset: the same data recalculated in terms of the nuclear spin temperature.

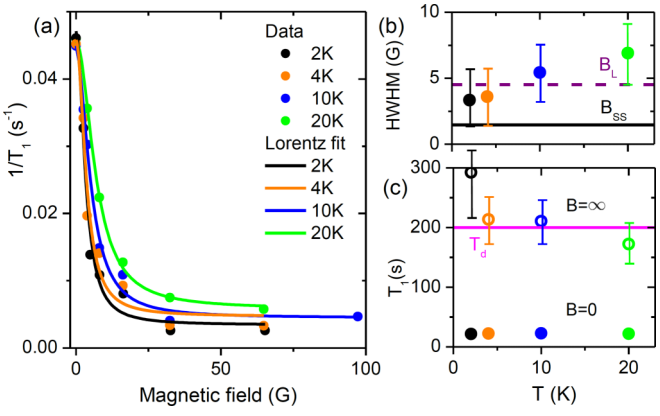

FIG. 3. (a) The NSS warm-up rate $1/T_1$ measured as a function of the applied magnetic field (symbols). Solid lines are fits to the Lorentz function. Values of the HWHM (b), T_1 at $B = \infty$ and $B = 0$ (c) extracted from the fitting procedure at different temperatures. (b) The solid line is the value of local field due to spin-spin interactions, B_{SS} [22], and the dashed line is the average value of the HWHM extracted from the data, which we interpret as B_L. (c) The solid line is a calculated value of the diffusion-limited relaxation time T_D.

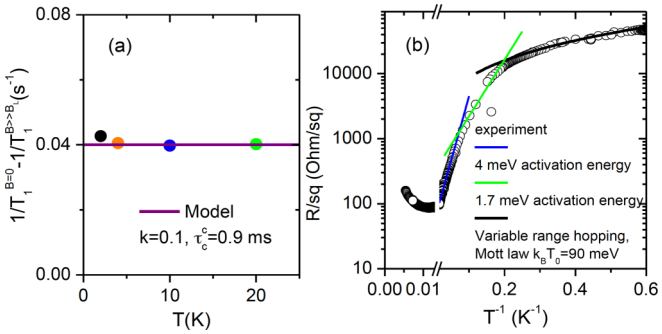

FIG. 4. (a) The enhancement of the NSS warm-up rate measured at different temperatures. The solid line is a fit to Eq. (13), with the only fitting parameter k/τ_c^c. (b) The electrical sheet resistance measured as a function of the inverse temperature. Solid lines are fits to the exponential behavior with two different activation energies, and the Mott law at $T \leqslant 5$ K.

is a huge, more than an order of magnitude enhancement of the NSS warm-up rate with magnetic field changing on the scale of several Gauss. The HWHM of this dependence, as well as zero- and strong-field limits of the field dependence vary only slightly over the studied temperature range [see Figs. 3(c) and 4(b)]. We present below the model that allows us to describe all these surprising results.

Theory. It is generally admitted that fast warm up of the NSS under the donor orbit via hyperfine interaction, followed by the spin diffusion towards donor sites is the main mechanism of the bulk NSS warm up [19,20,26]. Here we deal with another contribution to the bulk NSS warm-up, quadrupole interaction of nuclear spin with fluctuating electric fields. It results from the energy flux towards NSS via quadrupole interaction $\mathcal{F}_Q$:

$$\frac{1}{T_Q} = \frac{\mathcal{F}_Q}{\beta}(C_N)^{-1}, \tag{2}$$

where C_N is the heat capacity of the nuclear spin system [8]. Consider a small volume V, within which the fluctuating electric field E_f can be assumed spatially homogeneous. For GaAs and other piezoelectric semiconductors, the quadrupole Hamiltonian can be written in the following form:

$$\hat{H}_Q = -V(\vec{E}_f \vec{P}_Q), \tag{3}$$

where $\vec{P}_Q$ is the electric polarization related to the quadrupole magnetic moment. The components of $\vec{P}_Q$ are given by

$$P_Q^i = \frac{1}{V}\sum_{jk} \beta_Q \nu_{jk,i} \sum_n (Q_{jk})_n. \tag{4}$$

Here β_Q is the experimentally determined and isotope-dependent constant, eQ is the nuclear quadrupole moment, also isotope-dependent, e is the electron charge, $\nu_{jk,i} = 1$ if $i \neq j \neq k \neq i$, and zero otherwise. Therefore any electric field fluctuating at frequency ω induces an energy flux $\mathcal{F}_Q(\omega)$ towards nuclear spin system

$$\mathcal{F}_Q(\omega) = \frac{\omega}{2}\alpha''(\omega)E_f^2(\omega), \tag{5}$$

where $\alpha''(\omega)$ is the imaginary part of the generalized susceptibility. According to the fluctuation-dissipation theorem, $\alpha''(\omega)$ can be expressed through the power spectrum of the fluctuating part of $\vec{P}_Q$:

$$\alpha''(\omega) = V\frac{\omega\beta}{2}\langle \delta P_Q^2(\omega)\rangle. \tag{6}$$

To calculate the total quadrupole energy flux $\mathcal{F}_Q = \int_{-\infty}^{\infty}\mathcal{F}_Q(\omega)d\omega$ one needs to account for both electric field and quadrupole polarization fluctuation spectra. In the absence of illumination, the electric field fluctuates due to charge fluctuations induced by thermal activation of donor-bound electrons to the conduction band or, at lower temperatures, due to hopping to empty donors. Let us consider the simplest case, when the charge fluctuations are characterized by a single correlation time τ_c^c. It has a meaning of an average time needed to fill an empty donor site by phonon-assisted electron transition from the conduction band, or from a neighboring donor. In this case the time correlation function can be written as

$$\langle E_f(0)E_f(t)\rangle = \mathcal{L}E_a^2 e^{-t/\tau_c^c}. \tag{7}$$

Here E_a is the electric field at Bohr radius distance (a_B) from the charged donor position. The dimensionless coefficient $\mathcal{L}$ accounts for the averaging of the electric fields from the donor-bound electrons $\mathcal{L} = 2.5a_B^3 n_d^+$, where n_d^+ is the density of the charged donors [27]. Equation (7) is valid at time delays larger than the correlation time τ_c^{ph} of the fluctuating phonon field, which is usually much shorter than τ_c^c (typically τ_c^{ph} is in the picosecond range). Performing the Fourier transform of Eq. (7), we obtain the following expression for the power spectrum of the fluctuating electric field:

$$\langle E_f^2(\omega)\rangle = \mathcal{L}E_a^2\frac{2\tau_c^c}{1+(\omega\tau_c^c)^2}, \tag{8}$$

valid up to frequencies of the order of $2\pi/\tau_c^{ph}$. Therefore, the quadrupole energy flux can be expressed as

$$\mathcal{F}_Q = \frac{V\beta}{2\tau_c^c}\mathcal{L}E_a^2\int_{-\infty}^{\infty}\frac{(\omega\tau_c^c)^2}{1+(\omega\tau_c^c)^2}\langle \delta P_Q^2(\omega)\rangle d\omega. \tag{9}$$

Although the exact form of $\delta P_Q^2(\omega)$ is unknown, we can take into account that it spreads mainly in the frequency range $|\omega| < \gamma_N\sqrt{(B_L^2 + B^2)}$, where γ_N is the nuclear gyromagnetic ratio. Taking into account that $\tau_c^c \gg T_2 \sim 1/(\gamma_N B_L)$ we can replace the fraction under the integral by unity, so that

$$\mathcal{F}_Q = \frac{V\beta}{2\tau_c^c}\mathcal{L}E_a^2 2\pi\langle \delta P_Q^2\rangle. \tag{10}$$

Here $\langle \delta P_Q^2\rangle$ is the total squared fluctuation of quadrupole polarization. It can be calculated from quantum mechanical averaging over all nuclear spin states.

Nuclear heat capacity C_N can be written as [8,21,28]

$$C_N = \frac{N}{V}(C_Z + C_{SS} + C_Q), \tag{11}$$

where the three terms stand for the heat capacity associated with Zeeman, dipole-dipole, and quadrupole interaction, respectively, and N is the total number of nuclei in the volume V. The Zeeman part of the heat capacity is given by

$$C_Z = I(I+1)(\gamma_N \hbar)^2 B^2/3, \tag{12}$$

while spin-spin and quadrupole parts are usually expressed in terms of the corresponding effective fields B_{SS} and B_Q, so that $B_L^2 = B_{SS}^2 + B_Q^2$. Thus, we obtain the following formula for the nuclear spin warm-up rate:

$$1/T_Q = \frac{4\pi \mathcal{L}(eQ\beta_Q E_a)^2}{5(\hbar\gamma_N)^2\left(B^2 + B_L^2\right)\tau_c^c} \frac{4I(I+1)-3}{[8I(2I-1)]^2}. \tag{13}$$

One can see that this rate vanishes at strong magnetic field but can be important at low magnetic field. This results from the fact that nuclear heat capacity is strongly field dependent, while the the quadrupole energy flux is not. Thus, in the regime where hyperfine relaxation is limited by diffusion, the total warm-up rate is given by $1/T_1 = 1/T_Q + 1/T_D$, being determined by quadrupole contribution at $B < B_L$ and spin diffusion at strong field, as illustrated by Fig. 1(a).

Discussion. Let us now analyze our experimental data in the framework of the model. Hyperfine relaxation rate T_{hf} can be calculated as $T_{hf} = 1/(\omega_{hf}^2 \tau_c^s)$, where ω_{hf} is the nuclear spin precession frequency in the Knight field created by electron spin localized on the donor site, and τ_c^s is electron spin correlation time [8]. In this sample it amounts to $\omega_{hf} = 10^6$ rad/s, $\tau_c^s = 20$ ps [23], $T_{hf} = 0.05$ s, so that hyperfine relaxation of bulk nuclei is limited by the spin diffusion rate $1/T_D = 4\pi D n_d a_B$ [26], where $D = 10^{-13}$ cm^2/s is the nuclear spin diffusion coefficient [19]. With $a_B = 10$ nm we get $T_D = 200$ s, quite close to the experimentally obtained values at strong fields $B \gg B_L$ [solid line in Fig. 3(c)]. The spin diffusion rate does not depend on either temperature or magnetic field, so that all the observed variations should be related to the quadrupole mechanism.

To compare our model with the experimental results we use the values of $Q\beta_Q = 6\times10^{-15}$ cm and $\gamma_N = 1.5$ kHz/G averaged over all three GaAs isotopes, taking into account the abundance of each isotope [29]. Using $E_a = e/a_B^2 = 12$ kV/cm, $I = 3/2$, and $a_B = 10$ nm, we end up with three free parameters in Eq. (13): n_d^+, B_L, and τ_c^c. We assume that our sample has a considerable concentration of acceptors, so that at low temperature the concentration of charged donors $n_d^+ \approx k n_d$, where k is the compensation degree. It is easy to show that within this model, the HWHM of the $1/T_Q$ field dependence is exactly the value of B_L, while the height of the Lorentzian is $1/T_1^{(B=0)} - 1/T_1^{(B=\infty)}$. From this analysis we extract the enhancement of the NSS relaxation rate shown in Fig. 4(a) and $B_L = 4.5 \pm 2$ G, shown by the dashed line in Fig. 3(b). The spin-spin part $B_{SS} = 1.5$ G of the local field was calculated and deduced from experiments; its value is shown in Fig. 3(b) by a solid line [22]. The missing part of the local field ($B_Q \sim 4$ G) could be attributed to the quadrupole interactions due to uncontrolled strains or to some spin-spin interactions not accounted for in Ref. [22], such as the Dzyaloshinskii-Morya indirect exchange interaction [30]. Assuming the experimentally determined $B_L = 4.5 \pm 2$ G, we fit the field-induced enhancement of the warm-up rate by Eq. (13). The result is shown as a solid line in Fig. 4(a), with the fitting parameter $k/\tau_c^c = 110$ s^{-1}. This means, that for a reasonable compensation degree $k = 0.1$, the charge correlation time $\tau_c^c = 0.9$ ms. Therefore, our initial assumption $\tau_c^c > T_2$ is *a posteriori* confirmed. Thus, the quadrupole-induced relaxation model describes self-consistently the dramatic increase of the NSS warm-up rate at low magnetic fields. Equation (13) also suggests that the efficiency of this mechanism can be reduced in weakly compensated samples.

Since charge correlation time could be related to the resistivity, we also performed sheet resistance measurements in Van der Pauw configuration. The result is shown in Fig. 4(b), as a function of the inverse temperature. One can clearly see a nonmonotonous behavior, resulting from a combination of the polar optical phonon (above 80 K) and impurity scattering at lower temperatures [31,32]. In the latter case, we identify three regimes. The resistance between 50 and 20 K is governed by the thermal activation of bound electrons into the conduction band; the corresponding activation energy $\approx$4 meV is of the order of the donor binding energy. From 20 to 5 K electron hopping between donors takes over, with smaller value of the activation energy 1.7 meV. In this regime τ_c^c should be determined by the hopping between donors. Indeed, an estimation of the hopping transition times for typical donor pairs gives $\tau_c^c = 1$ ms at liquid helium temperatures [33]. Below 5 K, variable range hopping between donors takes over, giving rise to the Mott law behavior [34]. The resulting distribution of the hopping times within the impurity band should be exponentially broad; for this reason, charge fluctuations are expected to have a $1/f$ noise spectrum [35,36]. Thus, the simplified theory operating with a single charge correlation time may be not straightforwardly applicable at the lowest temperatures. Independent studies of the noise spectra and the generalization of the model to the case of a distribution of the charge correlation times should allow deeper insight in the mechanisms of the NSS warm-up rate enhancement at low magnetic field.

Conclusions. We have shown that the warm up of the NSS in bulk n-GaAs at low magnetic field is governed by a previously overlooked mechanism. This mechanism is mediated by the interaction of the quadrupole moment of the nuclei with slowly fluctuating electric fields, due to hopping of the electron charge, either into the conduction band, or across the impurity band. Our analysis suggests that a possible way to suppress the enhancement of the NSS warm up at low field could be found in a careful control and reduction of the compensation degree.

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