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We exploit the potential of the spin noise spectroscopy (SNS) for studies of nuclear spin dynamics in *n*-GaAs. The SNS experiments were performed on bulk *n*-type GaAs layers embedded into a high-finesse microcavity at negative detuning. In our experiments, nuclear spin polarisation initially prepared by optical pumping is monitored in real time via a shift of the peak position in the electron spin noise spectrum. We demonstrate that this shift is a direct measure of the Overhauser field acting on the electron spin. The dynamics of nuclear spin is shown to be strongly dependent on the electron concentration. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4922771]

Host lattice nuclear spins occupy a special place among spin systems in semiconductors. Indeed, their exceptional robustness to effects of environment opens up prospects to use nuclear spins for information processing.[1] On the other hand, they play major role in electron spin dynamics and decoherence.[2–4] Mechanisms of the nuclear spin relaxation in *n*-type semiconductors, even in the best studied like GaAs, remain rather poorly understood. Weak interaction of nuclear spins with light significantly complicates direct optical studies of nuclear spin dynamics. In general, nuclear spin state is detected via spin dynamics of charge carriers, in which case the carriers provide feedback on nuclei, often making difficult the interpretation of experiments.[5] An alternative method, so called two stage technique, was proposed in Refs. 6 and 7. In these experiments, nuclear spins are first cooled down by optical pumping, and in the second stage Faraday rotation induced by the Overhauser field is detected and used to trace the nuclear spin relaxation. Though this method shows a lot of promise for studies of nuclear spin relaxation, it gives relative values of nuclear fields, thus requiring complicated calibration procedures to determine the magnitude of nuclear polarisation.

Another technique that can be applied in the two stage arrangement is the spin noise spectroscopy (SNS). Primarily demonstrated on atomic systems,[8–11] this technique is widely applied to semiconductors and has been proved to be highly efficient.[12,13] The method is based on detection of the magnetization noise via fluctuations of the Faraday (Kerr) rotation of the laser beam polarisation upon transmission through (reflection from) the sample. If the laser frequency is tuned in the region of transparency, the detection is essentially non-perturbative. Only a significant increase of the probe beam power,[14,15] especially when the sample is placed inside a microcavity,[16] may result in noticeable light-induced perturbation.[15,17–20] Earlier experiments on electron spin noise (SN) in semiconductor nanostructures have revealed nuclear spin effects.[21] They appeared mainly as a broadening of SN spectra of localized charge carriers, caused by the frozen nuclear spin fluctuations.[22] It was also predicted theoretically that nuclear spin polarisation may result in drastic changes in SN spectra of localized electrons.[23]

In this Letter, we demonstrate that SNS can be used to investigate nuclear spin dynamics in *n*-doped semiconductors. We apply this technique to *n*-type GaAs layers embedded into a high-finesse microcavity to increase the detection sensitivity.[6,24] Two stage arrangement is implemented. First, nuclear spin polarisation is prepared by optical pumping with circularly polarised light, under magnetic field applied in Faraday geometry.[25,26] In the second stage, the dynamics of the initially prepared nuclear spin is monitored in the presence of an arbitrary transverse magnetic field B_x (Voigt geometry). The detection is achieved via electron SN spectra, recorded with time resolution of several seconds. A shift $\Delta\nu$ of the electron SN peak with respect to its unperturbed position, when nuclei are not polarised, is a direct measure of the Overhauser field $\boldsymbol{B}_N$ created by cooled nuclei $h\Delta\nu = \gamma_e B_N$. Here, γ_e is electron gyromagnetic ratio, well known for GaAs. Through this relation, SNS spectroscopy provides a direct measurement of B_N, with time resolution given by the SN spectrum acquisition time. The proposed method is applied to both metallic and insulating samples, revealing full potential of this technique, which allows to investigate nuclear spin relaxation in the presence of either donor-bound localized electrons, or mobile Fermi-edge electrons.

The studied samples are *n*-type GaAs epilayers embedded in a $3\lambda/2$ cavity characterised by the quality factor $Q \sim 10^4$ similar to those used in Refs. 6 and 24. Two similar samples with different doping levels were studied an insulating layer with $n \approx 2 \times 10^{15}$ cm^{-3} and a metallic layer with $n \approx 4 \times 10^{16}$ cm^{-3}. At these concentrations, electrons in *n*-type GaAs are characterised by similar spin relaxation times[27,28] of about 100 ns. However, at low temperatures

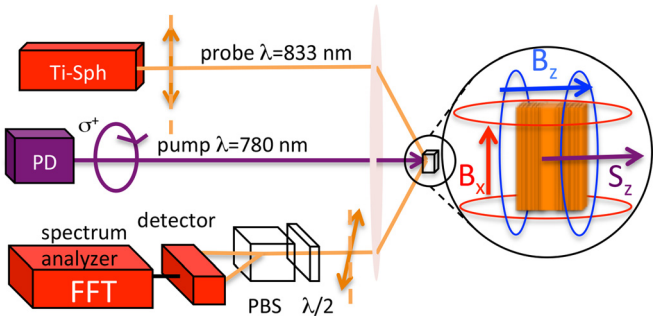

FIG. 1. Sketch of the two-stage experimental setup. Longitudinal magnetic field $B \uparrow\uparrow B_z$ is applied under pumping (stage 1). During measurement (stage 2) $B \uparrow\uparrow B_x$. In this pumping configuration $S_z \uparrow\uparrow B_z$ results in $B_N \uparrow\uparrow B_z$, so that $\Theta_N < 0$. Thus, at the measurement stage $B_N \uparrow\uparrow B_x$.

electrons in the insulating sample are bound to donors, while in the metallic sample they are delocalized and form a Fermi sea. The samples were mounted in a cold-finger cryostat ($T = 4$ K) surrounded by resistive coils for the application of magnetic field in both Faraday (B_z) and Voigt (B_x) geometries, as shown in Fig. 1.[29] Switching of magnetic field from B_z to B_x direction was typically realised in 1 ms and can be considered as adiabatic rotation.[30] In the first stage of experiment, nuclear spin polarisation was prepared by optical pumping with circularly polarised laser diode operating at 780 nm (just above Bragg mirrors stop-band) in the presence of the B_z. Typical pump power was $P_{pp} = 1$ mW. In the second stage, only linearly polarised probe beam resonant with the cavity mode provided by continuous wave (*cw*) Ti-Sapphire laser was sent onto the sample. Its wavelength is $\lambda \approx 833$ nm and typical power $P_{pr} = 0.25$ mW. The SN spectra of the probe beam reflected from the sample were recorded by means of a polarimetric optical scheme with the detector bandwidth up to 1 GHz and a FFT spectrum analyzer. The accumulation time of a SN spectrum could be reduced down to 1.5 s without affecting measurement accuracy. This defines temporal resolution of the experiments. Both pump and probe beams were focused on the spot of $\sim 30\,\mu$m diameter on the sample surface. Note also, that in the stage of preparation of nuclear polarisation, it was possible to replace the pump beam by intense (2 mW) circularly polarised beam at the probe energy that is in the GaAs transparency region. The latter could provide less efficient, but non-negligible nuclear spin cooling, probably due to residual absorption at the probe energy. Since the probe energy is close to the acceptor-conduction band transition in GaAs, one cannot exclude dynamic polarization of nuclei by resonantly excited spin-polarized holes.[31] The results shown here for metallic sample are obtained in this configuration.[32] We have carefully checked, however, that linearly polarised probe beam at typical power used during the measurement stage does not create itself any measurable modification in the SN spectrum. In contrast with the experiments on bulk GaAs, where the thickness of the layer where nuclei are efficiently polarised is very sensitive to the pump detuning with respect to the band gap, here the cavity layer is thin (≈200 nm). Thus, nuclear spins are polarized homogeneously over the entire cavity layer, irrespective of whether the structure is pumped above or below GaAs bandgap.

Figure 2 summarises SN measurements for both metallic and insulating samples. In the first stage of experiment, which precedes the SN measurement, nuclear polarisation was prepared by optical pumping in $B_z = \pm 120$ G. The duration of pumping was 5 min for metallic and 1 min for

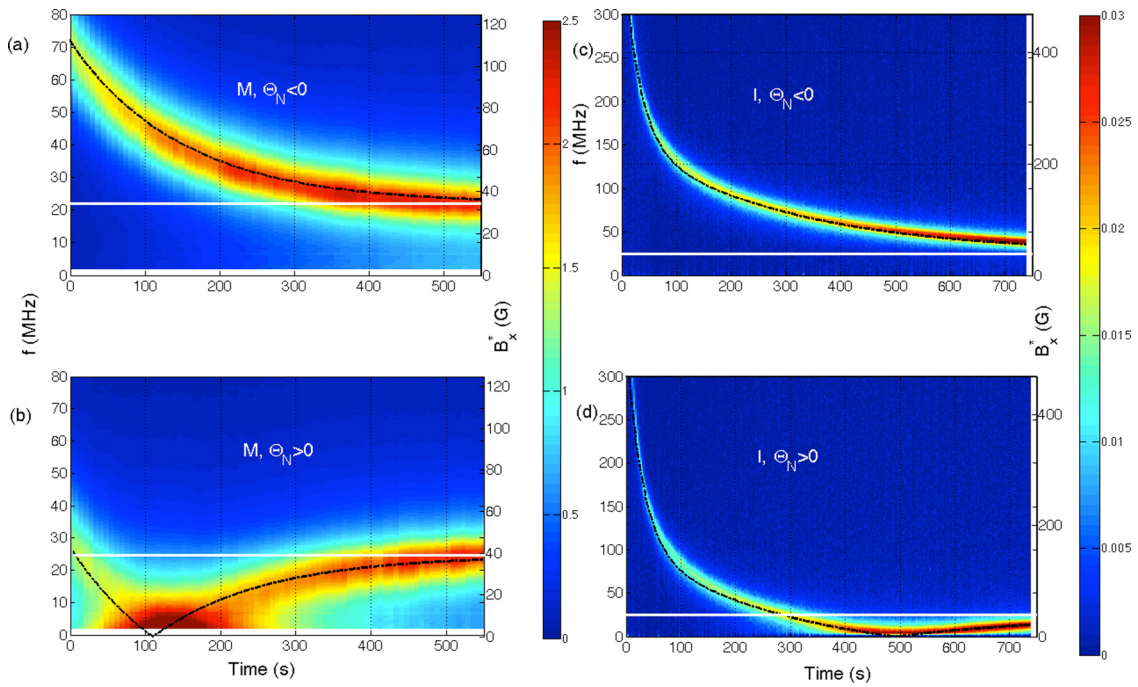

FIG. 2. Color maps of the time resolved spin noise spectra measured in metallic ((a) and (b)) and insulating ((c) and (d)) samples at $B_x = 40$ G. Color scale is given in the signal to shot noise ratio units, different for (a), (b) and (c), (d). In the first stage of experiment, nuclear spin polarisation was prepared under optical pumping in $B_z = 120$ G during 5 (1) min in metallic (insulating) sample. In (a) and (c), nuclei are prepared with negative spin temperature ($\Theta_N < 0$), so that Overhauser field is collinear with the external field. In (b) and (d), nuclei are prepared with positive spin temperature ($\Theta_N > 0$), so that Overhauser field is antiparallel to the external field. Right vertical scale provides the values of total field $B_x^* = B_N + B_x$ acting on the electron spin. Dashed lines are fits of the Overhauser field relaxation to the exponential ((a) and (b)) and bi-exponential ((c) and (d)) decay, solid lines indicate the values of the applied field.

insulating sample. Depending on the orientation of B_z with respect to the pump helicity, nuclear spin system is cooled down to either positive ($\Theta_N > 0$, $B_z \uparrow\downarrow S_z$) or negative temperature ($\Theta_N < 0$, $B_z \uparrow\uparrow S_z$).[25,33] Here, S_z is the electron spin polarisation, controlled by the pump helicity. At negative spin temperature, Overhauser field $B_N \uparrow\uparrow B_z$, while at positive temperature $B_N \uparrow\downarrow B_z$. At the second stage of the SN experiment, the pump beam is switched off, and the magnetic field is rotated to the perpendicular to light direction $B_x = 40$ G. The subsequent evolution of SN spectra measured every 1.5 s (7 s for metallic sample) is shown in Figure 2 as a colormap of the ratio between spin noise and shot noise powers.

Figure 2 is a key result of this work. It demonstrates the potentiality of SNS for studying nuclear spin dynamics. Indeed, the position of the SN peak in the spectrum is determined by the magnitude of the total field $B_x^* = B_x + B_N$ acting upon the electron spins, while its linewidth (at least for insulating sample) is determined by the distribution of nuclear fields. The gyromagnetic ratio for GaAs $\gamma_e \approx 0.64$ MHz/G allows us to directly relate the measured SN spectrum to B_x^*. The right axes in Figure 2 show the values of B_x^* obtained in our experiment. Solid lines indicate the contribution of the applied field B_x. One can see that the dispersion of nuclear fields is relatively small, which results in a narrow peak in the SN spectrum. Its dynamics reflects relaxation of the mean nuclear spin and the resulting Overhauser field B_N. Immediately after switching off the pump $B_N \gg B_x$, then B_N decreases. Relaxation of the nuclear spin polarisation is accompanied by the narrowing of the spectral line, indicating that the distribution of the nuclear fields narrows with decreasing B_N. Upper panels show the relaxation of nuclear spins prepared with $\Theta_N < 0$, so that B_x and B_N point in the same direction, while lower panels demonstrate the case of $\Theta_N > 0$, when these fields are anti-parallel. In the latter case, at some moment, namely, at $\sim$100 s for metallic and 500 s for insulating sample, the total field B_x^* crosses zero, as clearly seen in the plots. This corresponds to the so called compensation point, where $B_N = -B_x$. The striking difference between the time evolutions of the SN spectra measured for the two opposite signs of nuclear spin temperature is a tangible proof that this dynamics is due to nuclear spin, rather than any spurious thermal effect.

It is important to recall that the methodology of our experiment is a replication of the experiment devised by Kalevich, Kulkov, and Fleisher to demonstrate deep optical cooling of nuclei in semiconductors.[34] The important difference, however, is that the nuclear field is measured by SNS instead of the Hanle effect, i.e., depolarization of photoluminescence (PL) by a transverse magnetic field. The advantages of SNS with this respect are twofold. First, the range of measured nuclear fields is not limited by the width of the Hanle curve (determined by the electron spin lifetime[25]). Thus, nuclear spin dynamics can be studied in arbitrary transverse magnetic field. Second, in contrast with PL, but also with Larmor magnetometry based on Kerr or Faraday rotation,[35,36] SNS is a virtually non-perturbative method, which gives a possibility to study nuclear spin dynamics in the absence of photogenerated carriers.

To go further in the exploration of the SNS potential for the studies of nuclear spin dynamics, we compare the relaxation of nuclear spin in metallic and insulating sample. In metallic sample, the relaxation is, within the precision of our measurements, exponential. A fit to the monoexponential decay of the nuclear field is dashed-dotted curve in Figures 2(a) and 2(b). The characteristic decay time is $\tau \approx 150$ s. In the dielectric phase, it is apparently non-exponential; a reasonably good fit can be obtained using a bi-exponential decay with $\tau_1 \approx 30$ s, $\tau_2 \approx 300$ s, as shown in Figures 2(c) and 2(d). Qualitatively, this difference can be explained assuming that nuclear spin-lattice relaxation in the metallic semiconductor is mediated by itinerant Fermi-edge electrons via the Korringa mechanism,[37] while in the dielectric phase it is mediated by donor-bound electrons.[38] In the former case, the nuclear spin polarisation decays with equal speed at any spatial point. In the latter case, the polarization of nuclei under the orbits of donor-bound electrons decays much more rapidly than in the space between donors, where relaxation goes via nuclear spin diffusion towards donors, which play the role of killing centers. Such relaxation scenario results in two drastically different decay times for nuclear spin polarisation.[38]

It is instructive to compare two non-destructive approaches to the nuclear spin dynamics: the SNS proposed in this work, and Faraday rotation (FR), demonstrated in Ref. 6. Both methods can be applied to preliminarily cooled nuclei. In both cases, the wavelength of the probe is chosen in the transparency region of the studied semiconductor, so that probing goes without optical excitation of real electronic transitions, and therefore no perturbation is brought into the nuclear spin system. While the Overhauser-field-induced FR does not require presence of electrons to detect nuclear magnetization, the SN signal comes only from regions where resident electrons are present. For this reason, the two methods are complementary, making it possible to separate contributions of nuclei with stronger or weaker coupling to resident electrons in n-type structures. Combining the two approaches, a more detailed and quantitative study of the dependence of nuclear spin-lattice relaxation on donor concentration in *n*-type GaAs will be reported in our forthcoming publication.

In conclusion, we have demonstrated an original implementation of the SNS for studies of the nuclear spin relaxation in semiconductors. The method consists of two stages: (i) optical pumping of nuclear spins at first stage and (ii) measurement of the subsequent nuclear spin dynamics via electron SN spectra in the absence of the pump. Time resolution is limited by SN spectrum acquisition time, 1.5 s has been achieved in this work. The key strong points of SNS are the direct measurement of the Overhauser field in the presence of an arbitrary transverse field B_x, and its non-perturbative character. The possibility to perform measurements at low magnetic field is particularly precious, because this regime can hardly be accessed otherwise. It opens interesting prospects for studies of nuclear spin dynamics in *n*-type semiconductors, in particular, for the understanding of magnetic field and electron concentration dependence of the nuclear spin relaxation. Further development of SNS could include incorporation of the nuclear magnetic resonance

either during pumping or detection stage, in order to selectively probe nuclear species.

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