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Optical excitation effects on spin-noise spectroscopy in semiconductors

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The effects of laser intensity and laser wavelength on measurements of spin noise in bulk semiconductors are studied with an absorption-based optically excited impurity ionization model. The laser intensity and wavelength dependent electron spin relaxation time illustrates the gradual transition from a near nonperturbative measurement to a perturbative measurement. A strong relationship between the measured wavelength and intensity dependent absorption and the spin relaxation time is observed and is shown to fit well to a simple model. For semiconductors where spin noise has to be measured in the absorption regime, a spin relaxation time related to material properties rather than experimental conditions (e.g., laser intensity, laser wavelength, etc.) can be extracted from perturbative measurements in the limits of long wavelength and the low laser intensity.

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I. INTRODUCTION

Spin-noise spectroscopy (SNS) has been developed as a unique experimental technique to study spin states in atomic systems and semiconductors. In atomic systems, electron spin relaxation time and diamagnetic energy shifts have been studied with rubidium and ytterbium atoms.[1–3] Recently, SNS was adapted to the study of spin in semiconductors.[4–10] The electron spin relaxation time was measured by SNS in bulk *n*-GaAs (Ref. 4). With an optimized measurement technique improving sensitivity and efficiency, the effects of laser wavelength and temperature on the spin noise of n-GaAs were reported.[5] By using a mode-locked laser, spin noise in *n*-GaAs has been measured in the GHz range.[6] Moreover, the spin relaxation time in quantum confined structures like quantum wells and quantum dots has been investigated with SNS.[7,8] Through these measurements it was demonstrated that spin-noise spectroscopy can be used as an effective noncontact and nondestructive tool to map the three-dimensional doping density in semiconductors with a high spatial resolution and high accuracy, which has been a difficult task for traditional doping density measurement techniques like the Hall effect.[9] With the advantages of being nonperturbative, noncontact, and nondestructive, it is expected that SNS will find more applications in spintronics studies in the future.[10–15]

Compared with other experimental techniques utilized for spin measurements, like the Hanle effect that inevitably involves exciting the system in the optically absorbing region with near or above resonance excitation, SNS ideally uses a laser wavelength to probe the system with a below resonance excitation, giving the advantage of nonperturbative measurements.[1,4] Spin-noise measurements in atomic systems show little dependence on laser wavelength and intensity for the spin relaxation time. However, spin relaxation times measured in semiconductors by SNS show a strong dependence on laser wavelength and intensity. In SNS on quantum dots, due to the extremely low signal strength, the spin noise has to be measured in the absorption regime with near or above resonance excitation,[8] making it important to understand the effects of laser energy and intensity on SNS when absorption is present.

Several studies of spin noise in semiconductors as a function of electron density, magnetic field, temperature, laser wavelength, intensity, and interaction volume have been reported. Measurements as a function of electron density showed that a higher electron density near and above the metal-insulator transition gives a shorter spin relaxation time.[5,10,13] The magnetic field study gave the electron g factor.[4] The temperature study showed that the spin relaxation time decreases and the spin-noise power increases with increasing temperature.[5,10,13] The electron spin relaxation time of these structures has been measured by both SNS and the Hanle effect as a function of laser wavelength and intensity. Both spin-noise measurements and the Hanle effect measurements show a strong dependence of electron spin relaxation time on the wavelength and intensity of the probe laser, illustrating the undesired influence of subband gap absorption effects on the nominally “nonperturbative” measurements.[5,10] However, a full understanding of the effects of laser excitation on spin relaxation time as measured by SNS remains an interesting question.

In this paper the effects of laser excitation on spin-noise measurements are studied with a phenomenological optical excitation induced relaxation model incorporating the effects of both laser intensity and wavelength. First, the SNS is briefly introduced. Then the effects of laser intensity and laser wavelength on the electron spin relaxation time inferred from the width of the noise spectrum are quantitatively studied separately with the optical excitation induced relaxation model. At the end, a unified picture incorporating the effects of both laser intensity and laser wavelength is discussed with possible spin relaxation mechanisms.

II. SPIN-NOISE MEASUREMENT IN SEMICONDUCTORS

The experimental setup is shown in Fig. 1. A 45 degree linearly polarized laser beam passes through a 350-μm thick *n*-doped GaAs sample (doping density $\sim 1.8 \times 10^{16}\ \text{cm}^{-3}$) at a cryogenic temperature around 10 K with a pair of lenses. An external magnetic field is applied perpendicularly to the growth direction. The electron spin noise occurs as the spins precess around the external magnetic field. The spin flips create a fluctuating magnetic field along the laser propagation direction that leads to fluctuations in the energy levels of the spin states and corresponding fluctuations in the optical

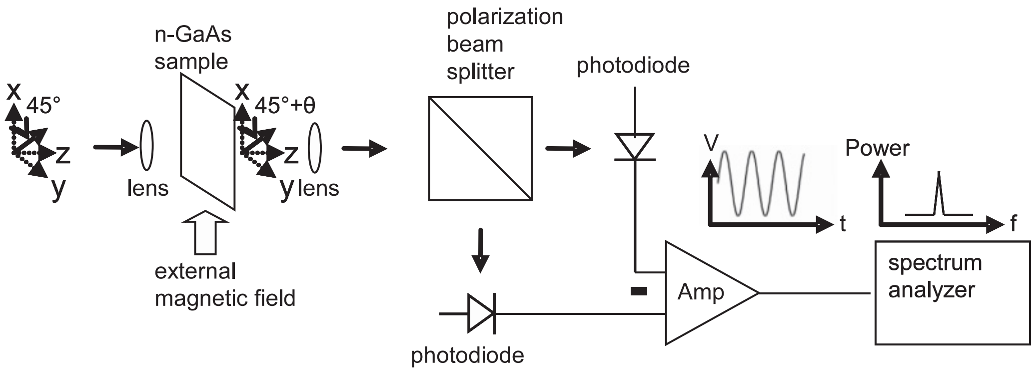

FIG. 1. Spin-noise measurement experimental setup. A 45 degree linearly polarized laser beam passing through a bulk n-GaAs sample under an external magnetic field, a polarization beam splitter, and being detected by a balanced detector and a spectrum analyzer.

energy level splitting. It is these fluctuations that lead to rapid fluctuations in the Verdet coefficient causing a broadening of the spectrum centered at the precession frequency as seen in a balanced detection of the corresponding Faraday rotation signal. The transmitted optical field is decomposed into two orthogonal, linearly polarized beams by a polarization beam splitter, and is detected with the balanced detector, as shown in the figure. Then the spin-noise signal is amplified by a rf amplifier and measured with a spectrum analyzer.

Due to inevitable noise in the measurement, the measured spectrum contains magnetic field dependent spin noise superimposed on a background of white noise including optical noise from the laser and electronic noise from the electronics. The background noise is characterized at zero magnetic field and is subtracted to obtain the spin noise. With a laser wavelength of 826 nm (1.501 eV) and laser intensity about 20 μW/μm^2, the measured spin-noise spectra at various magnetic fields are shown in Fig. 2(a) vertically shifted for clarity. Each spin-noise spectrum can be well fitted with a Lorentzian curve, such as the one shown in Fig. 2(b). The spin-noise width is the full width half maximum (FWHM) of the Lorentzian curve $f_{\rm FWHM}$, and the spin-noise power is the integrated power under the Lorentzian curve. The spin-noise width and power remain the same when the magnetic field changes. Peak frequencies f_p can be obtained from the Lorentzian curves. The magnetic field dependence of the peak frequency, shown in Fig. 2(c), gives an electron g factor value of about 0.4. The Lorentzian lineshape indicates a simple exponential decay with a spin relaxation time represented by the width of the Lorentzian spectrum. With the simple spin decay model, the spin relaxation time can be inferred from the measured spin-noise width.[9] The measured spin-noise width $f_{\rm FWHM}$ at various magnetic fields is about 7 MHz, as shown in Fig. 2(d), giving a spin relation time about 45 ns.

Beyond the intrinsic noise in the spin, the spin-noise power depends on detection related factors, like laser power on the detector and the optical-to-electrical signal conversion efficiency. To separate detector effects from the physics, an integrated spin noise, also called a Faraday rotation fluctuation, $\sqrt{\langle\theta_F^2(\lambda)\rangle}$, is defined and is extracted from the measured spin-noise power by normalizing the rms amplitude of the integrated spin-noise power to the dc voltage on the photodetector.[8] In atomic systems, the integrated spin noise is inversely proportional to the laser wavelength detuning and beam size,[1] as shown in Eq. (1)

$$\sqrt{\langle\theta_F^2(\lambda)\rangle} \sim \frac{1}{\Delta(\lambda)}\sqrt{\frac{L}{A}}\sqrt{f_e N_e}, \qquad (1)$$

where $\Delta(\lambda)$ is the detuning, L is the interaction length, A is the laser spot area, f_e is the fraction of the electron spins that are allowed to fluctuate, and N_e is the electron density.

In atomic systems the spin noise is measured with the laser wavelength in a transparency regime where the optical absorption is less than 5%.[1] The spin-noise width remains constant when the laser intensity and the wavelength change, indicating a nonperturbative measurement. In bulk semiconductors the spin noise has been measured in a regime where the absorption is between 10% to almost 100%. The spin-noise width changes significantly when laser intensity and wavelength change, indicating a perturbative measurement where both the laser intensity and the wavelength affect the spin relaxation time.[5,10] The optical absorption in the nontransparency regime is mainly due to impurities (the Urbach tail absorption) where absorption impurities absorb photons to generate optically excited ionized impurities and optically excited electrons.

To estimate the optically excited ionized impurity density, a simple linear model is used, as shown in Eq. (2). The amount of the optically excited ionized impurity is assumed to be linearly proportional to the laser power absorbed by the subband gap absorption, which has a strong dependence on laser wavelength and intensity. At a low excitation level the optically excited ionized impurity density is estimated from the absorbed laser power and the laser excitation volume

$$n_{ii}(\lambda,I) = n_{ii0} + \beta\frac{P_{\rm abs}(\lambda,I)}{V}\frac{1}{h\nu}\frac{1}{\gamma}, \qquad (2)$$

where $n_{ii}(\lambda,I)$ is the total ionized impurity density, n_{ii0} is the ionized impurity density without optical excitation, β is an excitation efficiency related to the probability of an absorbed photon leading ionization, $P_{\rm abs}(\lambda,I)$ is the laser wavelength and intensity dependent absorbed laser power, V is the laser excitation volume, $h\nu$ is the energy of the photon, and γ is the relaxation rate of optically excited ionized impurities. With an experimental model taking into account the laser intensity

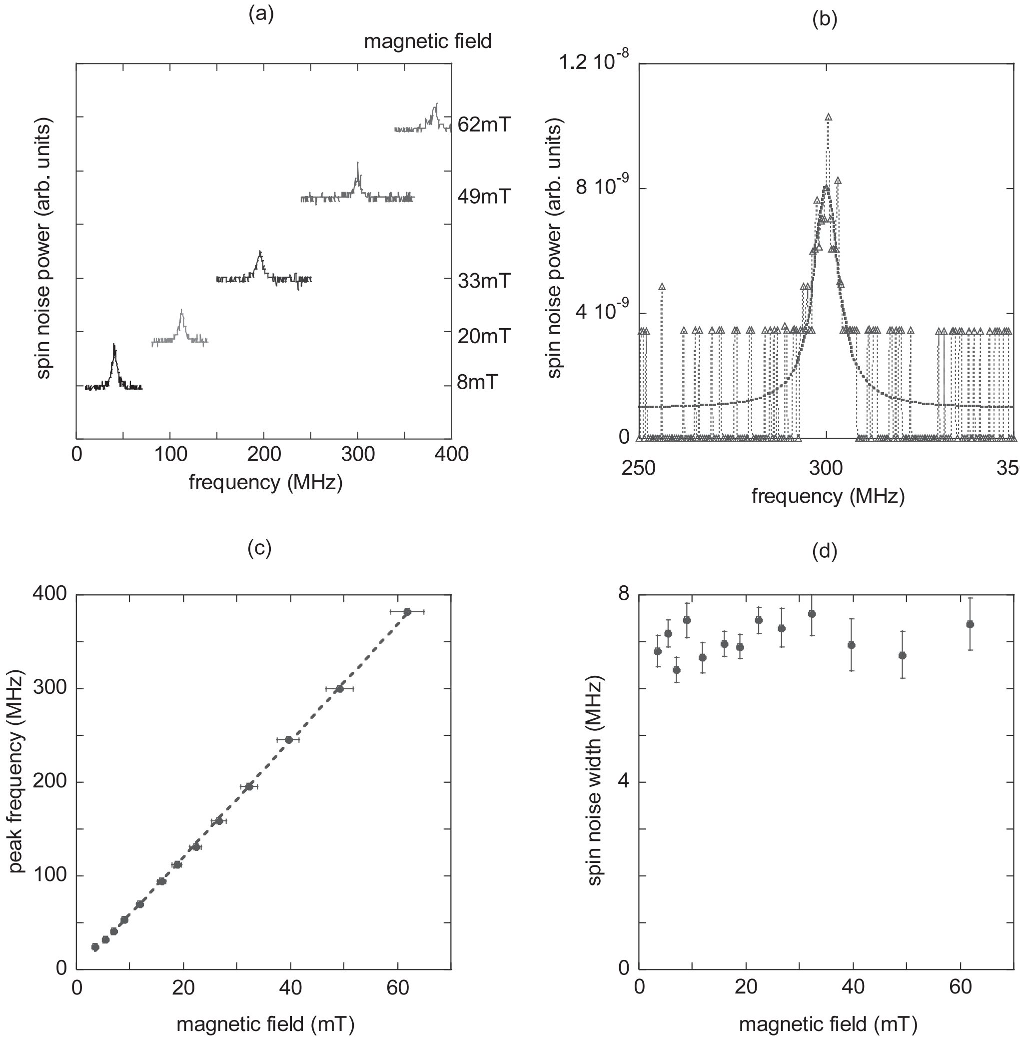

FIG. 2. Measured spin-noise spectra at different magnetic fields with a laser wavelength of 826 nm (1.501 eV) and laser intensity about 20 $\mu\text{W}/\mu\text{m}^2$. (a) Measured magnetic field dependent spin-noise spectra (vertically shifted for clarity). (b) Measured spin-noise spectrum at 49 mT with a Lorentzian fit (dashed curve). (c) Spin-noise peak frequency vs. magnetic field. (The dashed line is a linear fit.) (d) Spin-noise width vs. magnetic field.

and wavelength dependent ionized impurity amount and the laser excitation volume, the optically excited ionized impurity density can be estimated. Studying the correlation between optically excited ionized impurity density and spin relaxation time will help to understand the laser intensity and wavelength effects.

III. LASER INTENSITY AND WAVELENGTH EFFECTS

To study the effect of the laser intensity and wavelength on spin-noise measurements in bulk semiconductors, spin noise as well as the absorption are measured as a function of laser intensity and laser wavelength.

A. Dependence on laser intensity

To study the effect of laser intensity, the laser wavelength was fixed at 826 nm (1.501 eV), a wavelength where spin noise over a large laser intensity range can be measured. The laser intensity was changed from 27 to about 1100 $\mu\text{W}/\mu\text{m}^2$ by adjusting the laser power with a fixed laser spot size. Figure 3(a) shows the measured laser intensity dependent spin-noise spectra. The peaks are not at the same frequency due to magnetic field strength variations in each measurement in our experiments (the positioning of the field could not be precisely controlled). Figure 3(b) is the absorption coefficient over the laser intensity with an insert showing the laser intensity dependent transmission and absorption. Figure 3(c) shows the laser intensity dependent spin-noise width with an insert showing the integrated spin noise. At high laser intensities the spin-noise spectra could deviate from a Lorentzian curve due to complications caused by a weak thermal effect (the sample temperature increased about 2 to 3 degrees at the highest laser intensity we measured). Strictly speaking, the laser intensity effect we studied involves a temperature effect. However, temperature dependent studies with low laser intensities showed that the 2 to 3 degree temperature increase at 10 K only increases the spin-noise width no more than 15%.[5]

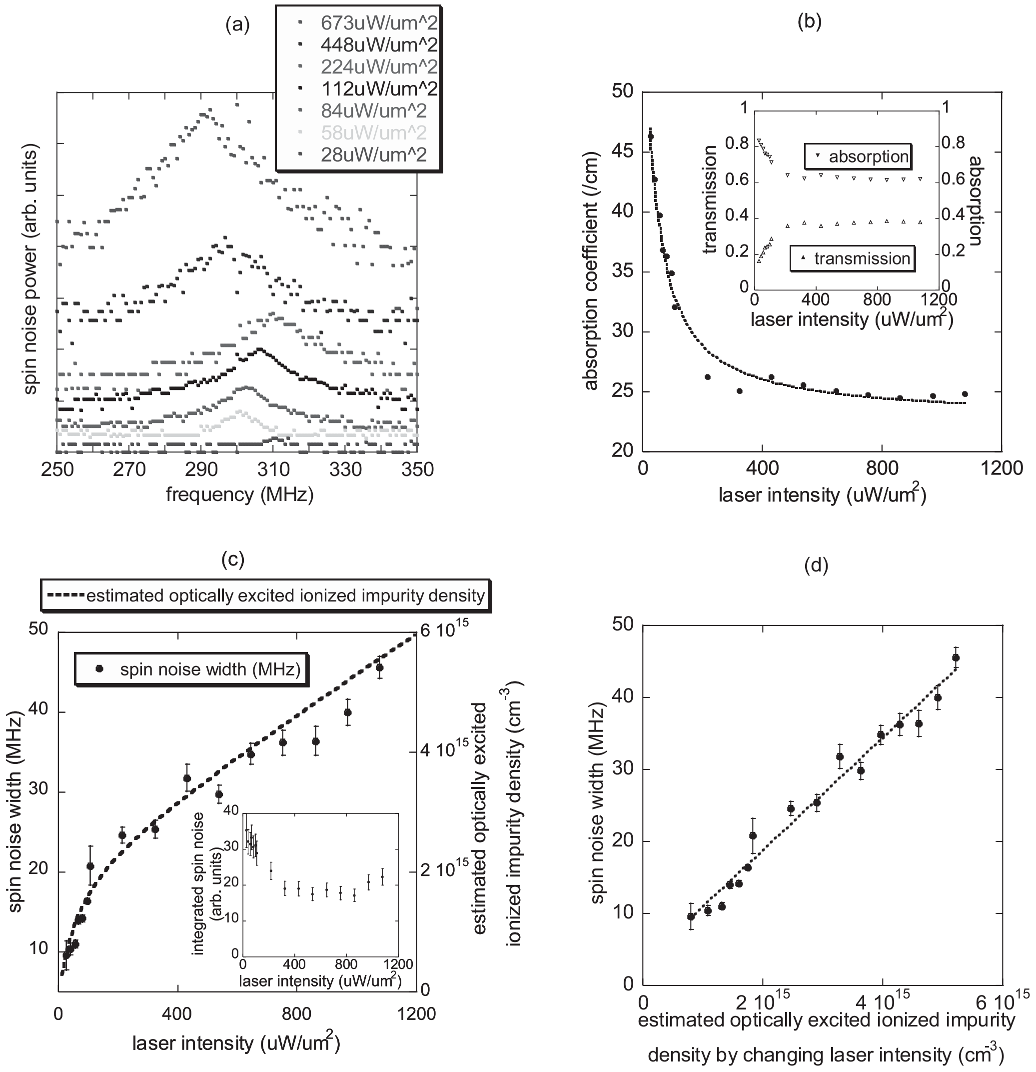

FIG. 3. Laser intensity effect on spin-noise measurements. (a) Laser intensity dependent spin-noise spectra measured with laser wavelength 826 nm (1.501 eV) at 10 K (vertically shifted for clarity). The peaks are not at the same frequency due to magnetic field strength variations in each measurement in our experiments. (b) Sample absorption coefficient vs. laser intensity. The dotted curve is a fit with a two-level model. The insert shows the sample transmission and absorption vs. laser intensity. (c) Spin-noise width and estimated optically excited ionized impurity density vs. laser intensity. The insert shows the integrated spin noise vs. laser intensity. (d) Spin-noise width vs. optically excited ionized impurity density.

The amount of change we observed, six fold, is mainly from the intrinsic physics associated with the laser excitation rather than an artifact due to the slight change in the temperature.

To understand the laser intensity effect, the absorption coefficient is calculated from the measured transmission, as shown in Fig. 3(b). In the low intensity regime, the absorption coefficient is high and the absorbed power increases rapidly when the laser intensity increases. In the high intensity regime, the absorption coefficient gets saturated and the absorbed power increases slowly when the laser intensity increases. The physics of the impurity absorption is beyond the scope of this study, but the laser intensity dependent absorption coefficient fits well with a two-level model as Eq. (3)[16]

$$\alpha(I) = \frac{\alpha_0}{1+\frac{I}{I_{\text{sat}}}} + \alpha_{ns}, \tag{3}$$

where α_0 is the zero intensity absorption coefficient, I is the laser intensity, I_{sat} is the saturation intensity when the absorption drops to half, and $\alpha_{ns}$is a nonsaturating absorption coefficient. The curve fitting gives a zero intensity absorption coefficient $\alpha_0 = 45 \pm 6\,\text{cm}^{-1}$, a saturation intensity $I_{\text{sat}} = 33 \pm 8\,\mu\text{W}/\mu\text{m}^2$, and a nonsaturating absorption coefficient $\alpha_{ns} = 23 \pm 1\,\text{cm}^{-1}$. The insert of Fig. 3(c) shows that the spin-noise signal strength drops when the absorption coefficient saturates.

With the two-level saturation model, the laser intensity along the laser beam path can be calculated. Based on the simple linear excitation model (i.e., the absorption is nonlinear in the intensity, but the optically excited ionized impurity density is assumed to depend linearly on the absorbed power) mentioned above, an upper limit on the optically excited ionized impurity density can be estimated by first fitting the width of the noise spectrum as a function of incident power.

We leave as free parameters both the fraction of the absorbed photon that leads to ionized impurities (i.e., quantum efficiency for ionization) from the saturating absorption as well as from the nonsaturating absorption. With an overall scaling factor and constraining the upper limit of the quantum efficiencies to be unity, we found a smooth chi-square surface with a minimum for the saturating term of unity and the quantum efficiency for the nonsaturating term of 0.07. This fit is shown in Fig. 3(c). Note that this fit leads to a finite noise width at zero intensity, presumably due to the intrinsic ionized impurity density and other impurity independent noise terms. This is a simple linear relation as seen by the parametric plot (noise as a function of estimated ionized impurity density) in Fig. 3(d). In the calculation, the relaxation rate of optically excited ionized impurities of 23 ns is used.[17] An upper limit of the excitation efficiency β of 3% is used, based on that the optically excited ionized impurity at the high laser intensity is expected to be lower than the doping density. Uncertainty or variation of the excitation efficiency βwill only affect the scaling factor for the estimated optically excited ionized impurity density, without affecting the number for the finite noise width at zero intensity. We note that because of the crudeness of the model, we have assumed the absorbed power along the laser propagation direction is constant leading to a uniform ionized impurity density. This is clearly an oversimplification because of the sample thickness, but does not impact the qualitative picture we are using.

In the low intensity regime, the optically excited ionized impurity density increases rapidly when the laser intensity increases since the absorption is not saturated. In the high intensity regime, the optically excited ionized impurity density increases slowly when the laser intensity increases since the absorption is saturated. The linear fit indicates a strong correlation between the spin-noise width and optically excited ionized impurity density. Extrapolation to zero gives a lower limit of the spin-noise width of about 3 MHz, indicating a spin relaxation time of 105 ns. A similar laser intensity effect is also observed by changing the laser spot size with a fixed laser power.[10]

B. Dependence on laser wavelength

To study the dependence on the laser wavelength, the laser intensity was fixed at about 140 μW/μm^2, and the laser wavelength was tuned from 818 (1.505 eV) to 850 nm (1.465 eV). Figure 4(a) shows the dependence of the noise spectrum on the laser wavelength. As in the earlier figure the peaks are not at the same frequency due to the difficulty of setting the magnetic intensity for each measurement in our experiments.

To understand the wavelength dependence, the absorption coefficient was calculated from the measured transmission, as shown in Fig. 4(b). The absorption in this wavelength range arises from the Urbach tail, mainly caused by subband-gap absorption in bulk semiconductors due to impurities.[19–21] For the purpose of describing the laser wavelength dependence with a formula fitting with the experimental data, the laser wavelength dependence was well modeled with a Lorentzian.[22] To show the similarity to the data in Refs. 5 and, 10 the inset in Fig. 4(c) shows the integrated spin noise as a function of laser energy.

With the Lorentzian model, the optically excited ionized impurity density is estimated using the same model above used to analyze the data in Fig. 3. With the measured absorption coefficient at each wavelength, the intensity along the depth is calculated. With the corresponding laser power absorbed, the optically excited ionized impurity density is numerically calculated, as shown in Fig. 4(c). Figure 4(d) plots the spin-noise width vs. estimated optically excited ionized impurity density, where a linear fit indicates a linear correlation between the spin-noise width and the optically excited ionized impurity density. Extrapolation of the linear fitting gives about 7.6 MHz, a lower limit of the spin-noise width at long wavelength limit corresponding to the laser intensity used in the measurements, indicating a spin relaxation time of 41 ns.

IV. DISCUSSION

The measurement of spin noise is emerging as a potentially useful and relatively nonperturbing method for determining the spin relaxation time in semiconductors, an issue of increasing importance for some spintronic and quantum information applications. The numbers reported here for the spin noise are in general agreement with those in the literature,[4,5,10,13] though longer spin lifetimes (∼500 ns) have been reported by other techniques.[23,24]

It is important to consider potential systematic errors that could modify the numbers or the observed behavior. For these measurements, the sample had an absorption length comparable or shorter than the sample thickness for the highest absorbing regions of the measurement. In addition, the optical field has the usual transverse Gaussian intensity profile. Hence, both of these lead to spatial inhomogeneity in the absorbed power as a function of length and radial position in the optical field. The inhomogeneity becomes more complicated at intensity levels close to or in excess of saturation. To evaluate the impact of this, we estimated the absorbed power and corresponding optically excited ionized impurity density with a model including the saturation effects along both longitudinal and transverse directions based on the laser intensity dependent absorption curve we measured. The difference between the more complete model and the simple model is about 7%. From this, we conclude that the noise numbers could be smaller by perhaps up to 20%, which does not affect the qualitative agreement between the data and the model. We also note that the estimated ionized impurity density is based on numbers extracted from the literature. With the current data set, exact numbers are not possible. However, the uncertainty in these numbers effects only scaling and does not impact the fitting of the data or conclusions about the success of the simple model.

To understand the effect of the optical excitation on the spin relaxation time measured by SNS in semiconductors, it is necessary to look into the physics of the spin relaxation mechanism in this system. There are mainly three spin relaxation mechanisms affecting the electron spin relaxation time in bulk semiconductors,[25] the Elliot-Yafet (EY) mechanism,[26] the D'yakonov-Perel (DP) mechanism,[27] and the Bir-Aronov-Pikus (BAP) mechanism.[28] Optical excitation may generate electron holes, and electron-ionized impurities

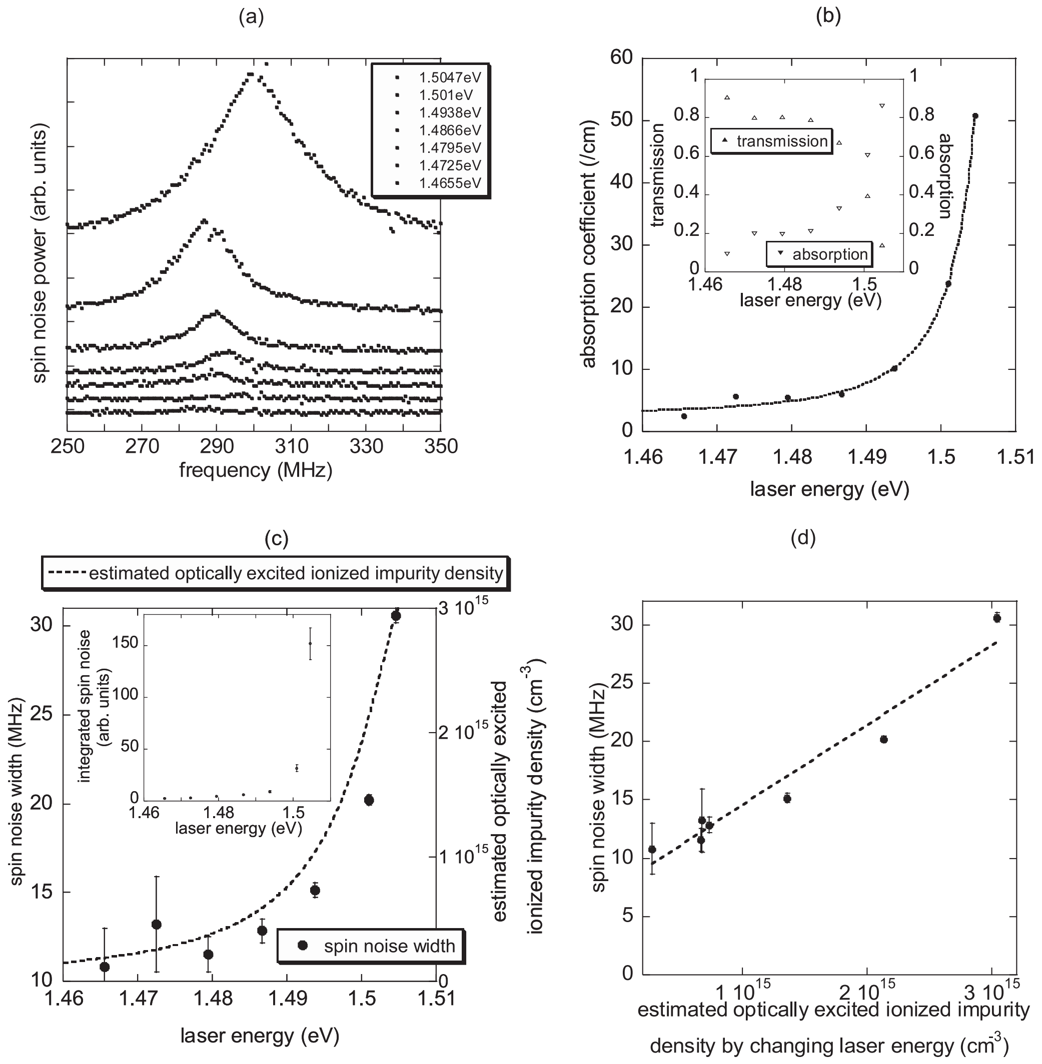

FIG. 4. Laser wavelength effect on spin-noise measurements. (a) Laser wavelength dependent spin-noise spectra measured with a laser intensity of 140 μW/μm^2 at 10 K (vertically shifted for clarity). The peaks are not at the same frequency due to magnetic field strength variation in each measurement in our experiments. (b) Sample absorption coefficient vs. laser energy. The dotted curve is a fit with the Lorentzian model. The insert shows the transmission and absorption. (c) Spin-noise width and estimated optically excited ionized impurity density vs. laser energy. The insert shows the integrated spin noise vs. laser energy. (d) Spin-noise width vs. optically excited ionized impurity density.

as well. There could be multiple physical mechanisms that cause spin relaxation following optical excitation.

Crooker *et al.*[10] showed that the BAP mechanism may play a role in causing spin relaxation when the optical excitation energy is close to 1.515 eV in GaAs, the energy of the exciton. Here, the spin relaxation is dominated by interactions with the resultant hole produced by optical excitation.

A second path that can lead to the decreasing spin relaxation time is via impurity ionization under the optical excitation. The impurity absorption peaks are at 1.491 and 1.493 eV in *n*-GaAs (Refs. 17 and 18). In the laser energy range in spin-noise measurement, 1.46–1.505 eV, impurity ionization should dominate. The optically excited impurity ionization may affect the measured spin relaxation time through two different mechanisms. One is the DP mechanism, where the electron spin relaxation time is increased by the scattering of the electron with ionized impurities. However, studies have shown that in lightly doped *n*-GaAs increasing doping density gives a shorter spin relaxation time since the Fermi energy increases with increasing doping density.[29,30] If the optically excited impurities affect the Fermi energy in the same way as thermally excited impurities, optical excitation will reduce the spin relaxation time.

Another mechanism that the optically excited ionized impurities may affect the spin relaxation time is the recombination of ionized impurities with electrons.[29,31] Under a weak optical excitation, the measured electron spin relaxation time is mainly the electron spin relaxation time of a steady state of thermally excited electrons. Under a strong optical excitation, the measured spin relaxation time is the electron spin relaxation time of a steady state of all electrons including the thermally excited electrons and the optically excited electrons. The optically excited ionized impurities may recombine with electrons. When the electron spin relaxation time (41–103 ns as measured here) is longer than the electron-impurity recombination time (around 23 ns for *n*-GaAs with doping density of 8.7×10^{14} cm^{-3}) (Ref. 17) the measured spin relaxation time will be reduced under strong optical excitation.

The simple model presented in this paper implies a linear dependence on optically excited ionized impurity density. But the data are not adequate to suggest one relaxation mechanism over another.

Given the clear dependence on the probe field intensity and wavelength, laser based measurements of the intrinsic material spin relaxation time should be performed in the transparency regime, where excitation is minimized as spin-noise studies on atomic systems have done.[1] However, experimentally it is hard to implement spin-noise spectroscopy in semiconductors in the transparency regime due to the long wavelength range of the Urbach tail. Indeed, for quantum dots it is reported that the spin noise has to be measured in the absorption regime due to the weak signal in these systems.[8] An indirect but possible solution is to measure the spin-noise width over a two-dimensional coordinate system including both laser wavelength and laser intensity. This is time consuming given the number of data points that might be needed; but it is expected that an intrinsic material spin relaxation time will be obtained by extrapolating the two-dimensional laser wavelength and intensity dependent spin-noise width in the limit of long laser wavelength and the zero intensity. This extrapolation method applies to the spin relaxation time measurement on bulk semiconductors based on the Hanle effect as well since the probe beam is within the same wavelength and intensity range as the spin-noise measurement.[10]

V. SUMMARY

We have presented data examining the role of laser intensity and wavelength on spin-noise measurements in bulk doped semiconductors. The laser wavelength and intensity dependent electron spin relaxation time was related to the optical absorption, which then, through a simple model, was used to estimate the optically generated ionized impurity density. The data showed a simple correlation between the electron spin relation rate and the ionized impurity density, consistent with other experiments. The laser wavelength and intensity dependent electron spin relaxation time illustrates the gradual transition from a near nonperturbative measurement to a perturbative measurement. However, additional data would be needed to obtain the fully nonperturbative relaxation rate.

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