

Light-induced Knight shifts in GaAs/Al_xGa_{1-x}As quantum wells

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The coupling between quantum-confined electron spins in semiconductor heterostructures and nuclear spins dominates the dephasing of spin qubits in III/V semiconductors. The interaction can be measured through the electron-spin dynamics or through its effect on the nuclear spin. Here, we discuss the resulting shift of the NMR frequency (the Knight shift) and measure its size as a function of the charge-carrier density for photo-excited charge carriers in a GaAs quantum well.

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I. INTRODUCTION

The spin of electrons in semiconductors represents a degree of freedom that has recently been discovered as a possible handle for high-speed electronic devices¹ and other spintronic devices.² In a similar context, the electron spin has been proposed as a suitable degree of freedom for storing and processing quantum information.³⁻⁵ In both cases, the performance of these devices is strongly affected by the hyperfine interaction of the electrons with the nuclear spins, which shifts the electron-spin energy levels, changes the Larmor frequency, and is, under some experimentally relevant conditions, the limiting factor for the dephasing of electronic spins in III/V semiconductors.⁵⁻⁷ In cases where the nuclear spin carries the quantum information, the interaction with an electronic spin is often used as a control interaction.³ Such schemes rely, therefore, on a precise knowledge of and possible means for control of the hyperfine coupling.

In this paper, we investigate the hyperfine coupling between conduction-band electrons and nuclear spins in GaAs quantum wells. Since the GaAs conduction band consists of *s*-type orbitals, the hyperfine coupling is dominated by the Fermi-contact term, which is proportional to the electron-spin density at the site of the nucleus.

Hyperfine interactions in III/V semiconductors are never observed as resolved splittings. The individual couplings are relatively small ($\Delta E/h < 1$ MHz), and individual electrons have comparable couplings with approximately 10^5 – 10^6 nuclear spins. The observable average effect of the nuclear spins on the electronic spin is therefore that of an effective magnetic field, which is called the nuclear field. It has been observed, e.g., as a shift of the Hanle curve⁸ or as a change of the electronic Larmor frequency.⁹

Similarly, the electron spins modify the dynamics of the nuclear spins. Since the relaxation time of the electrons is always fast on the time scale of the individual hyperfine interaction, the nuclear spins do not respond to a single electronic spin, but to a time-averaged effective interaction. The fluctuations of the hyperfine coupling drive the exchange of polarization between the electronic and nuclear spin system. This polarization transfer has been used to increase the polarization of the nuclear spins.^{10-14,18} In addition, the time-averaged hyperfine interaction shifts the nuclear Larmor frequency by an amount that is proportional to the electron

density, the hyperfine coupling strength, and the electron-spin polarization. This shift of the nuclear resonance frequency, which is known as the Knight shift, has been studied extensively in metals, where it was first discovered.¹⁵ In semiconductors, the shifts are much smaller, due to the low electron density. First results were obtained in gray tin (α -Sn),¹⁶ where the small band gap results in relatively large shifts at room temperature and above. In this case, the measured Knight shifts could be used to obtain information about the band structure. Similarly, Knight shifts can be used to study metal-to-insulator transitions (see, e.g., Ref. 17).

In quantum wells, quantum dots, and other semiconductor heterostructures, the electronic wave function is modified by confinement. Using the proportionality of the Knight shift, it has been proposed to use it to map the excited-state electron wave function of a quantum well,¹⁸⁻²⁰ possibly with atomic resolution.²¹ However, the relatively small electron-spin density and the small number of nuclear spins in quantum-confined heterostructures makes the measurement of Knight shifts a serious problem.²⁰

Some of the optically detected NMR techniques that have been used to overcome the sensitivity issues of NMR in individual heterostructures such as quantum wells and quantum dots cause severe broadening of the NMR transitions,^{18,19} thereby obscuring the relatively small Knight shifts. These problems can be avoided with a more recently introduced experiment,²² which uses optical pumping to increase the nuclear spin polarization as well as optical detection, but combines them with pulsed radiofrequency (rf) excitation of the nuclear spins to avoid power broadening: during detection, the nuclear spins precess freely, in the absence of an rf field. The Fourier transform of the optically detected free induction decay, therefore, shows the natural width of the NMR transitions and is ideally suited to measure small resonance shifts with high precision and accuracy. At the same time it provides high sensitivity, yielding single-shot spectra of individual quantum wells. Here, we use these capabilities to measure the Knight shift from optically generated conduction-band electrons.

The paper is organized as follows. In Sec. II, we review the fundamentals of the generation of spin-polarized photo-excited electrons in the conduction band and calculate the expected Knight shift. Sections III and IV describe the experimental setup and present the experimental results, and in

Sec. V, we summarize and draw conclusions.

II. LIGHT-INDUCED KNIGHT SHIFT

We consider the following system: A single electron spin, which is excited into the conduction band by the absorption of circularly polarized light, couples to an external magnetic field via the Zeeman interaction and to a large number of nuclear spins via the hyperfine coupling. The relevant Hamiltonian is then

$$\mathcal{H} = \mathcal{H}_{ze} + \mathcal{H}_{hf} + \mathcal{H}_{zn} = B_0 \mu_B g^* S_z + \sum_i a_i \vec{S} \cdot \vec{I}^i + B_0 \gamma I_z^i, \quad (1)$$

where we use the conventional choice of coordinate system (z axis parallel to the external magnetic field B_0). \vec{S} is the electron spin operator, \vec{I}^i represent the nuclear spins, a_i are the hyperfine coupling constants for the i th nuclear spin, B_0 is the strength of the magnetic field, μ_B is Bohr's magneton, and g^* is the effective g factor, modified by the confinement potential of the quantum well.

The hyperfine coupling strength is proportional to the electron density at the site of the nucleus. Paget *et al.*¹⁸ calculated the maximum value of the coupling constants for all three isotopes in GaAs as $a^{75\text{As}}(r=0) = -160$ kHz, $a^{69\text{Ga}}(r=0) = -133$ kHz, and $a^{71\text{Ga}}(r=0) = -169$ kHz, assuming a Bohr radius of the exciton, a_0^* , of 10 nm. For an exponentially decaying envelope of the exciton wave function, the coupling constant decreases exponentially with the distance from the center of the exciton, $a_i(r) \propto e^{-2r/a_0^*}$.

We consider only the case where the interaction of the electron spin with the external magnetic field is significantly stronger than the hyperfine coupling. The x and y components of the hyperfine coupling term are then nonsecular. While they remain relevant for the exchange of polarization between electron and nuclear spin, their effect on the static Knight shift is negligible.

Since the lifetime of an exciton is short compared to the time scale of the nuclear spin dynamics, the effective coupling strength experienced by the nuclear spins has to be evaluated as the time average over the generation, motion, and decay of the excitons. The average number of excitons is (at most) $\bar{N}_e = P_{\text{abs}} \tau_e / h\nu$, where P_{abs} is the absorbed optical power in the region of interest, τ_e is the exciton lifetime, and $h\nu$ is the photon energy. For an absorbed power of 1 mW and an exciton lifetime of about 300 ps, the average number of excitons in the illuminated volume becomes, therefore, $\bar{N}_e = 1.2 \times 10^6$. We calculate the average hyperfine coupling acting on the nuclear spins, assuming that the excitons are homogeneously distributed throughout the illuminated volume V_{il} from the ratio of the average exciton density $\bar{n}_e = \bar{N}_e / V_{\text{il}}$ to the electron density at the center of a single exciton,

$$\bar{a} = a(r=0) \bar{n}_e V_e, \quad (2)$$

where $V_e = 4\pi \int_{r=0}^{\infty} r^2 e^{-2r/a_0^*} dr = \pi (a_0^*)^3$ is the volume of a single exciton.

If the spins were static, this coupling could be observed as a splitting in the NMR spectrum. However, since the (spin) lifetime of the electrons is much shorter than the inverse of the coupling strength (in frequency units), one actually observes the time average of a spectrum that hops between the two positions. The resulting shift is

$$\langle \delta\nu \rangle_t = \bar{a} \langle S_z \rangle_t, \quad (3)$$

where $\langle S_z \rangle_t$ is the time-averaged z component of the electron spin.

III. EXPERIMENTAL DETAILS

To obtain the high sensitivity required to measure NMR spectra from individual quantum films, we had to combine optical pumping of the nuclear spin system with optical detection of the precessing nuclear spins.²² Furthermore, we had to be able to modify the laser irradiation during the experiment. For that purpose, the laser beam (from a titanium sapphire ring laser) was split into two parts, which could both be controlled independently with an acousto-optic modulator. The beams were recombined on a polarizing beam splitter and then passed through a $\lambda/4$ retardation plate. Depending on which AOM was switched on, the laser light was thus polarized σ^+ or σ^- behind the $\lambda/4$ plate. The light was then focused onto a $37 \mu\text{m}$ $1/e$ diameter spot on the sample.

Our sample is a multiple-quantum-well structure grown by molecular-beam epitaxy (MBE) containing 13 quantum wells with thicknesses from 2.8 to 38 nm. The quantum wells are separated by 39 nm $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers. The sample was mounted on the cold finger of a helium flow cryostat and the temperature was kept at 2.8 K. The laser wavelength was tuned to 812.3 nm, to resonantly excite the 20 nm quantum well. A 1.367 T magnetic field was applied at an angle of 70° from the direction of the laser beam.

The photoluminescence from the sample was collected with a lens, analyzed with a circular polarizer, and measured with an avalanche photodiode (APD) with 10 MHz bandwidth. Nuclear spin transients were excited by a radiofrequency (rf) pulse with a duration of $6 \mu\text{s}$ and a frequency of 9.971 MHz, close to the resonance frequency for ^{75}As . As described in an earlier paper,²² the resulting signal is closely related to the free induction decay (FID), and the NMR spectrum can be recovered by Fourier transformation of the time-domain signal.

Figure 1 shows a typical spectrum. The splitting is due to quadrupole interaction of the $I=3/2$ nuclear spin, which would be absent in an ideal GaAs crystal²³ and indicates a distortion of the quantum-well sample.²⁴

Since the Knight shifts are relatively small, we measured them in a two-dimensional NMR experiment, using different electron-spin densities during the evolution and detection periods. During the evolution period, we varied the polarization and intensity of the exciting light and compared the resulting frequency with the precession frequency during the detection period, where the laser irradiation was applied with constant circular polarization and constant intensity.

Figure 2 shows the sequence of optical and radiofrequency fields applied to the sample during an experiment.

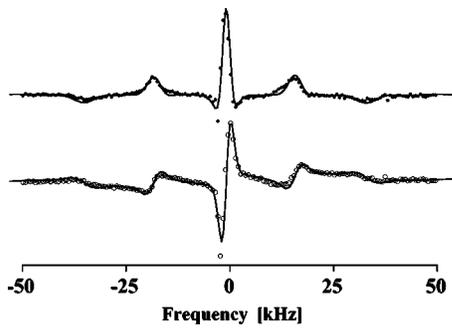


FIG. 1. Typical single-shot NMR spectrum from a 20-nm-wide QW after 160 sec optical pumping. Dots represent experimental data and solid lines the theory. The upper trace shows the absorption signal, the lower the dispersion.

Each experiment starts with irradiation of σ_+ light to create nuclear spin polarization. The first radiofrequency pulse converts the longitudinal to transverse magnetization, which precesses for a time t_1 . During this evolution time, we applied optical irradiation with either σ_+ or σ_- polarization and variable intensity to modify the spin polarization of the conduction electrons. The phase that the nuclear spin coherence acquires during this time is a measure of the precession frequency, which includes the effect of the Knight shift. The second and third radiofrequency pulses store the magnetization along the magnetic field direction while the polarization of the optical radiation is switched. The optical FID (OFID) is recorded during t_2 . The sequence is then repeated after a delay of 30 s.

As usual in a 2D NMR experiment, the evolution time was incremented systematically, from 10 μ s to 1280 μ s in steps of 10 μ s. τ_m was kept constant at 50 ms. The resulting data were stored in a two-dimensional data set and Fourier transformed with respect to the two time variables t_1 and t_2 to generate a two-dimensional spectrum with frequency variables ν_1 and ν_2 . Quadrature detection in the ν_1 direction was obtained by adding the signals of two spectra with identical timing but with a 90° phase shift for the last two pulses.

IV. KNIGHT SHIFT

Figure 3 shows an example of a resulting 2D spectrum. The dashed line marks the diagonal of the spectrum, while

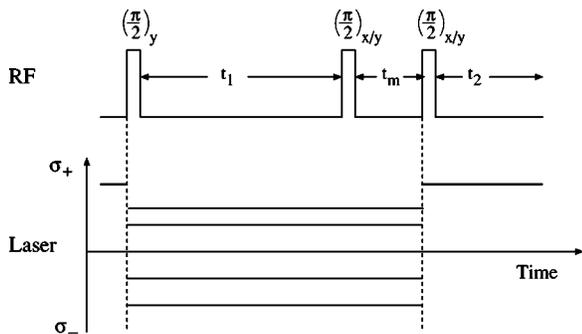


FIG. 2. Timing of the experiment. The upper line shows the three radiofrequency pulses, which control the nuclear spin magnetization. The second trace shows the intensity of the circularly polarized light, upward for σ_+ and down for σ_- .

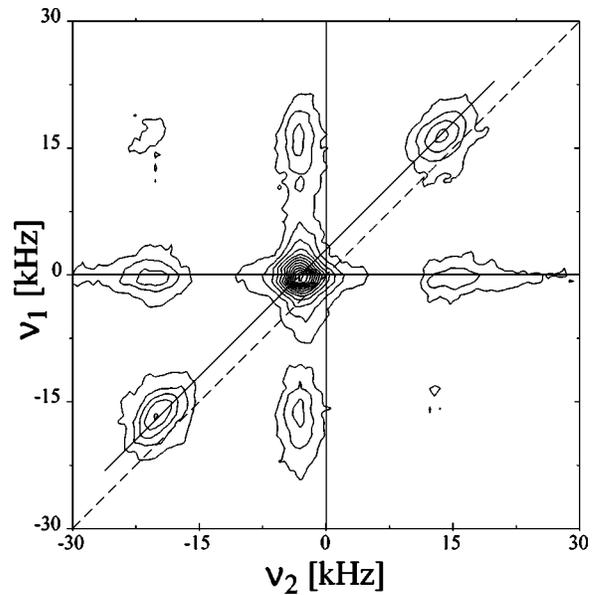


FIG. 3. 2D spectrum from a 20-nm-wide QW obtained with excitation with 42 mW σ^+ light during detection and 21 mW σ^- light during evolution. The horizontal axis corresponds to the precession frequency during detection, the vertical one to the evolution frequency.

the solid line connects the main resonance lines in the spectrum. The vertical displacement between these lines represents the difference in the Zeeman frequencies of the nuclear spins as the optical irradiation changes from σ^- to σ^+ .

This shift should depend on the average electron-spin polarization. We varied the electron-spin density through the intensity of the optical pump light and the spin polarization by switching between left and right circular polarization.

Figure 4 shows three representative spectra that were obtained for σ^- (bottom), no (center), and σ^+ optical irradiation

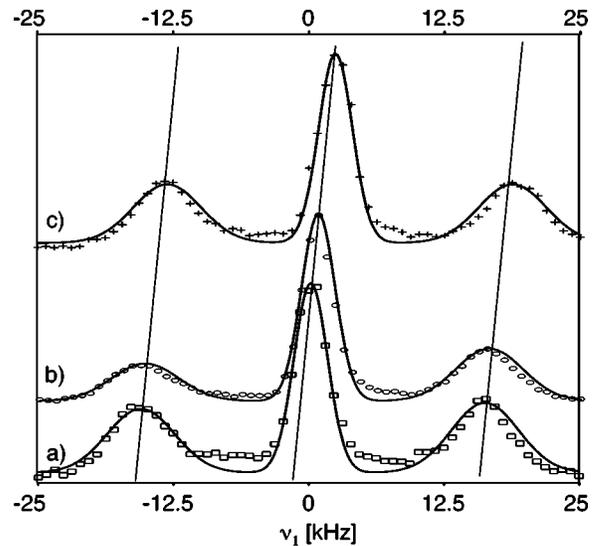


FIG. 4. Variation of the Knight shift with the optical irradiation. Each spectrum represents a slice through the 2D spectrum parallel to the ν_1 axis. Optical irradiation during evolution: (a) σ^- 20 mW, (b) none, and (c) σ^+ 39 mW.

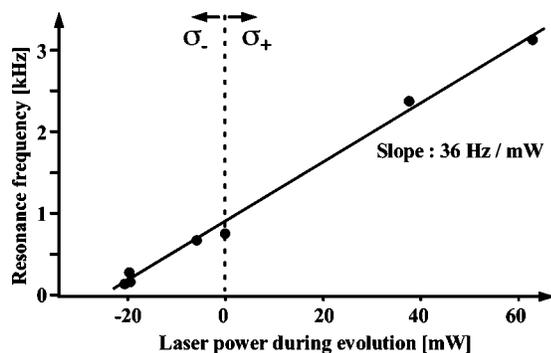


FIG. 5. Variation of the nuclear Larmor frequency with polarization and intensity of the laser light.

during t_1 . As indicated by the straight lines, the position of all three resonance lines changes in the same way, in direct proportion to the density of photoexcited electrons. This is consistent with the behavior expected for a light-induced Knight shift.

For a more systematic evaluation, we fitted the center transition in each spectrum to a Gaussian resonance line and measured the change in precession frequency during the evolution time. For this analysis, we used only the central transition, which is not broadened by quadrupolar couplings and therefore allows a more precise measurement of the center frequency. Figure 5 summarizes the resulting line positions for different polarizations and laser intensities. The variation shows the expected linear dependence on the number of spin-polarized electrons. From the slope of the linear fit, we calculate a Knight shift of 36 Hz/mW of laser radiation.

To check the plausibility of these data, we use Eqs. (2) and (3), which predict a line shift of

$$\langle \delta\nu \rangle_t = a(r=0) \bar{n}_e \overline{V_e \langle S_z \rangle}_t. \quad (4)$$

We calculate the time-averaged electron-spin polarization $\langle S_z \rangle_t = \langle S_0 \rangle \cos \theta = 0.3 \times \cos 70^\circ = 0.1$ from the measured po-

larization of the photoluminescence $\langle S_0 \rangle$ and the angle $\theta = 70^\circ$ between the laser beam and the magnetic field. Per milliwatt of *absorbed* light, we then expect a shift of about 3 kHz. Since about 50% of the incident light is reflected at the windows of the cryostat and the surface of the sample, this is consistent with the observed shift if 2.5% of the light entering the quantum well is converted into conduction-band electrons.

V. CONCLUSION

The hyperfine interaction between the spins of charge carriers in quantum-confined semiconductor heterostructures and the nuclear spins in the material affects the dynamics of the electronic and nuclear spins in different ways. Here, we have concentrated on the time-averaged effect of the electron spin on the nuclear spins. This effect can be described as a shift of the resonance frequency, which is proportional to the electron density and the polarization of the electron spins.

Since the small electron densities make it difficult to observe the Knight shift in semiconductors, we used an experimental setup that was designed to optimize the sensitivity while minimizing unwanted perturbations of resonance frequency, thus providing optimal resolution.

Our samples were nominally undoped, and the experiments were conducted at liquid-helium temperature. The resulting Knight shift was therefore dominated by the photoexcited electrons. A rough estimate of the expected shifts is in good agreement with the experimental data.

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