Optically pumped NMR: Revealing spin-dependent Landau level transitions in GaAs

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(Received 20 July 2010; published 30 August 2010)

We show that high-resolution optically pumped NMR (OPNMR) studies can reveal spin-dependent optical transitions between valence- and conduction-band Landau levels in bulk semiconductors such as GaAs. The OPNMR signal intensity exhibits oscillations as a function of pump photon energy that evolve with magnetic field. In contrast to standard polarized magnetoabsorption measurements, OPNMR is sensitive to the *polarization* of the photoexcited electron spins (i.e., the *difference* between spin-up and spin-down electron populations rather than the *sum*). This allows one to clearly resolve the spin dependence of optical transitions that might normally be obscured in conventional magnetoabsorption studies. The data are in good agreement with theoretical calculations of the transitions from the spin-split light-hole Landau levels in the valence band to the conduction-band Landau levels of GaAs.

DOI: 10.1103/PhysRevB.82.085209 PACS number(s): 71.70.Di, 31.30.Gs, 76.60.−k, 78.20.Ci

I. INTRODUCTION

Knowledge of the electronic structure of crystalline solids is vital to understanding their electronic, transport, and optical properties. Traditionally, magnetic fields have played an important role in determining band structure, with cyclotron resonance or magnetoabsorption being used for semiconductors. For spintronics and other applications, it is particularly desirable to understand the spin-dependent band structure. Optically pumped NMR (OPNMR) is the combination of light excitation for optical pumping of semiconductors with radio-frequency (rf) (NMR) detection of nuclear spins. OPNMR results in significant sensitivity enhancements over typical thermal polarizations of the nuclear spin system, and signals are localized to regions of a sample where the light is absorbed. OPNMR has been used to study diverse phenomena including optically generated Overhauser effects,[1] dipolar spin order,[2] the manifestation of skyrmions,[3] the fractional quantum Hall ground state,[4] and electron spin polarization.[5]

In OPNMR, semiconductors are optically pumped using circularly polarized photons to polarize the conduction-electron spins.[6] These spin-polarized electrons subsequently polarize the nuclear spins through a Fermi-contact hyperfine interaction, substantially enhancing the NMR signal strength.[7–10] The sign and magnitude of the OPNMR signal will depend on the average electron spin polarization $\langle S_{opt}(\hbar\omega)\rangle$ created through optical pumping.[6,11] As such, OPNMR is inherently sensitive to the *sign* and *magnitude* of the electron spin polarization and is thus very different from (but complementary to) magnetoabsorption, which is sensitive to the net optical absorption and which does not explicitly depend on the electron spin polarization. As an illustrative example, it is well known that purely circularly polarized light does not generate 100% polarized electrons in bulk GaAs: Due to the optical selection rules, σ^- circularly polarized light will excite "spin-up" electrons from heavy-hole states in the valence band, *and* will also excite "spin-down" electrons from light-hole valence states. On average, the oscillator strength of the light-hole transitions is smaller than that of the heavy holes by a factor of 3; however, this ratio can vary significantly at particular photon energies in a magnetic field due to the formation of spin-split Landau levels

OPNMR studies to date have shown a connection between the macroscopic features in the optical absorption, $\alpha(\hbar\omega)$, where $\hbar\omega$ is the photon energy, and the characteristic variation in OPNMR signal intensity for optical excitation *below* the bandgap ($\hbar\omega < E_g$).[1,12,13] Above E_g, oscillations in the OPNMR intensity were observed as a function of photon energy; however, a detailed analysis of their physical origin was lacking. Such phenomena had been reported previously in several bulk semiconductors and in quantum wells.[8,14–16] In one report on $Al_xGa_{1-x}As/GaAs$ quantum wells, the periodic features in the OPNMR intensity were attributed to formation of Landau levels[7,8] but it was suggested that such phenomena would not be observable in bulk samples due to the impurity concentration.[17] In a recent study, we attributed the oscillatory features in OPNMR of GaAs to Landau levels[18] but without a corresponding match to specific transitions between the levels, for which theoretical calculations are critical.

Here we show that OPNMR, in conjunction with theoretical calculations and optical magnetoabsorption, can be a powerful tool for understanding the band structure and spin-dependent optical transitions in semiconductors. We demonstrate that the weaker transitions from the light-hole Landau levels dominate the OPNMR signals. We report on OPNMR spectra of ^{69}Ga spins in bulk semi-insulating GaAs polarized by a narrowband laser in two different external (B_0) magnetic fields of 4.7 tesla (T) and 7.0 T. These experiments combined with magnetoabsorption allow one to measure the complicated spin-split valence-band Landau levels, resolving details of the electronic spin structure of the valence bands of GaAs.

II. EXPERIMENT

Experiments were carried out on semi-insulating GaAs obtained from ITME, Warszawa, Poland (sample character-

istics: growth direction [100], thickness 400 μm, mobility 5630 $cm^2/V\,s$). The samples were each mounted in a home-built single-channel transmission line probe inside a continuous-flow cryostat (Janis-200 Supertran), to maintain the sample temperature at 6 ± 0.2 K.

The ^{69}Ga OPNMR experiments were recorded at a Larmor frequency of 48.09 MHz (B_0=4.7 T) and 70.77 MHz (B_0=7.0 T). The spectra were collected using pulse sequences consisting of a saturating rf train (SAT), followed by a period of continuous-wave laser irradiation, τ_L (120 s), a single $\frac{\pi}{2}$ pulse, and signal acquisition (ACQ): SAT$-\tau_L-\frac{\pi}{2}-$ACQ. The saturating train consisted of 50 $\frac{\pi}{2}$ rf pulses separated by 1 ms each. The data acquisition and processing were performed using a Tecmag Apollo console. The τ_L period of 120 s was selected to attenuate effects due to spin diffusion while still providing sufficient signal-to-noise ratios for analysis.[11] The experimental procedure used also includes recording a reference spectrum in a single shot without laser irradiation, which we term the "Boltzmann" signal. The purpose is to record the resonance frequency of the ^{69}Ga nuclei in the sample at 6 K populated by thermal processes and not due to coupling to the optically oriented electrons or to the laser.

A tunable continuous-wave frequency-stabilized Ti:Sapphire laser (Coherent 899–21, ~500 kHz linewidth) pumped by a 532 nm solid-state diode laser (Spectra Physics Millenia X) was used as an optical excitation source. The wavelength of the Ti:Sapphire output was measured with an Ocean Optics HR-2000 spectrometer (resolution of 0.035 nm). The linearly polarized output from the laser was converted to σ^+ or σ^- polarized light using a quarter-wave retarder, centered at 825 nm (retardation tolerance is within $\frac{\lambda}{50}$ for the range used in these experiments). In all the experiments presented here, the laser power was kept constant at 2.5 W/cm^2. The sample was irradiated with the incident beam parallel to the external magnetic field, and the beam diameter was 4 ± 1 mm. The laser was blocked only during the saturating rf train and between acquisitions; it was on during the pulse sequence and during acquisition.

III. RESULTS AND DISCUSSION

^{69}Ga and ^{71}Ga OPNMR signals of GaAs are characterized by spectra whose sign changes with the helicity of light used, with positively phased signals when exciting with σ^- circularly polarized light and negatively phased signals with σ^+ circularly polarized light [Figs. 1(a) and 1(b), respectively]. Further, the OPNMR signal intensity exhibits complicated oscillations as a function of above-gap photon energy ($E_g > 1.521$ eV), shown for ^{69}Ga in Figs. 1(c) and 1(d) for σ^- and σ^+ excitation. These oscillations change their peak-to-peak spacing as a function of magnetic field, as well, and OPNMR measurements at 4.7 and 7.0 T were reported.[18] Similar oscillations have been observed previously in several bulk semiconductors and quantum wells but the physical origin for the observed behavior in bulk samples was previously unknown.[8,14–16]

We have found that effects of the spin-split valence-band Landau levels can be observed in the bulk OPNMR spectra.

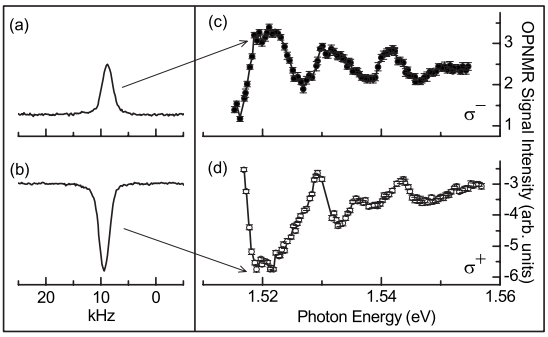

FIG. 1. Representative ^{69}Ga OPNMR spectra at 4.7 T recorded for (a) σ^- and (b) σ^+ excitation (λ=815 nm) showing the phase inversion of the NMR resonance between light helicities and the ^{69}Ga OPNMR signal intensity (integrated peak area) as a function of photon energy for (c) σ^- and (d) σ^+ excitation.

By combining our experiments with theoretical calculations (Fig. 2), we can identify the specific transitions responsible for the oscillations. Because the OPNMR signals depend explicitly on the spin polarization of the conduction electrons rather than on the sum of transitions that pump spin-up and spin-down electrons (as is the case for polarized magnetoabsorption), we show that the additional structure associated with light-hole-to-conduction-band transitions can be resolved by the OPNMR signals which otherwise may be difficult to detect in magnetoabsorption.

In some cases (e.g., InSb) where there is a very large g factor,[19] magnetoabsorption can be used to determine the spin splittings in the conduction band;[20] yet, in GaAs and many other materials, the g factor is small. Consequently, the conduction bands are nearly spin degenerate (shown in blue and labeled C0, C1, C2 in Fig. 2). However, in GaAs as seen in Fig. 2, spin splittings in the valence band are large enough

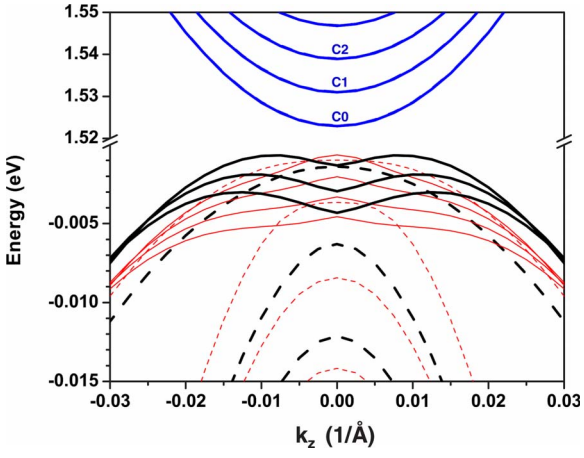

FIG. 2. (Color online) Calculated spin-split valence-band and conduction-band Landau levels in GaAs at 4.7 T. Black (thick) lines correspond to heavy holes, red (thin) lines to light holes, and blue lines (labeled C0, C1, C2) to conduction band levels. Solid lines are for spin-up and dashed lines are for spin-down states. These assignments are only approximate due to band mixing. Only the lowest few Landau levels of each type are shown. Spin-up and spin-down states for the conduction band are nearly degenerate and are not resolved in this figure.

to be resolved by OPNMR. OPNMR is sensitive to the sign of the polarization (i.e., the direction of the nuclear spin polarization with respect to the external magnetic field), thereby providing information on the spin-dependent electronic states and optical transitions of GaAs.

^{69}Ga OPNMR signals were measured for both light helicities as a function of excitation energy in order to observe the effect of optical pumping at 4.7 T [σ^- and σ^+, Figs. 1(c) and 1(d), respectively]. It is evident from Fig. 1 that the OPNMR intensity is dependent on both excitation energy and light helicity, in a manner similar to oscillations observed in magnetoabsorption measurements.[21]

To theoretically predict and identify the oscillations in the OPNMR signal intensity several steps are needed. First, we calculate the energy levels of bulk GaAs in a magnetic field. Determination of the energy levels and eigenfunctions is achieved by numerically solving a set of coupled effective mass equations based on the Pidgeon-Brown model.[22,23] In the effective mass theory, we retain conduction electrons, heavy, light, and split-off holes. The resulting Hamiltonian factors into a set of 8×8 matrices, each of which corresponds to a specific magnetic manifold and is described in detail elsewhere.[24,25] Results of our electronic structure calculation, using known band parameters for GaAs (Ref. 26) are displayed for the spin-split valence and conduction bands at 4.7 T in Fig. 2.

After the electronic levels and states are determined, we then calculate the optical absorption from the imaginary part of the dielectric function ϵ_2. Using Fermi's golden rule, and the notation in Ref. 24, we find that for undoped systems,

$$\epsilon_2(\hbar\omega) = \frac{e^2}{\Lambda^2(\hbar\omega)^2} \sum_{n,\nu;n',\nu'} \int_{-\infty}^{\infty} dk_z |\hat{e}\cdot\vec{P}_{n,\nu}^{n',\nu'}(k_z)|^2 \times \delta(\Delta E_{n',\nu'}^{n,\nu}(k_z) - \hbar\omega), \quad (1)$$

where n and n' are the Pidgeon-Brown manifold numbers, ν and ν' label the eigenvectors, $\Lambda=\sqrt{\hbar c/eB}$ is the magnetic length, and $\Delta E_{n',\nu'}^{n,\nu}(k_z)=E_{n',\nu'}(k_z)-E_{n,\nu}(k_z)$ is the transition energy. The interband optical matrix elements are,[24]

$$\hat{e}\cdot\vec{P}_{n,\nu}^{n',\nu'}(k_z) = \sum_{m,m'} a_{n,m,\nu}^{*}(k_z) a_{n',m',\nu'}(k_z) \times \langle\Phi_{N(n,m)}|\Phi_{N(n',m')}\rangle\langle m|(\hat{e}\cdot\vec{P})|m'\rangle, \quad (2)$$

where $\hat{e}$ is the unit polarization vector of the radiation incident along the z axis, $a_{n,m,\nu}$ are the complex expansion coefficients of the envelope wave functions, and $\Phi_{N(n,m)}$ are the orthonormal harmonic oscillator functions.

Because GaAs is a relatively wide band-gap material, we find that the conduction band states are almost completely pure spin states (the mixing of the opposite spin state through the $\vec{k}\cdot\vec{p}$ interaction is less than 2%). As a result, if we restrict the sum in Eq. (1) over the final conduction-band states of a given spin, we can calculate the absorption for a given spin type. For example, using either σ^+ or σ^- light, $\alpha_\uparrow$ represents the amount of absorption that creates spin-up electrons, $\alpha_\downarrow$ represents the amount of absorption that creates spin-down electrons, and the total magnetoabsorption is given by $\alpha=\alpha_\uparrow+\alpha_\downarrow$. The conduction-band spin polarization is therefore $(\alpha_\uparrow-\alpha_\downarrow)/(\alpha_\uparrow+\alpha_\downarrow)$.

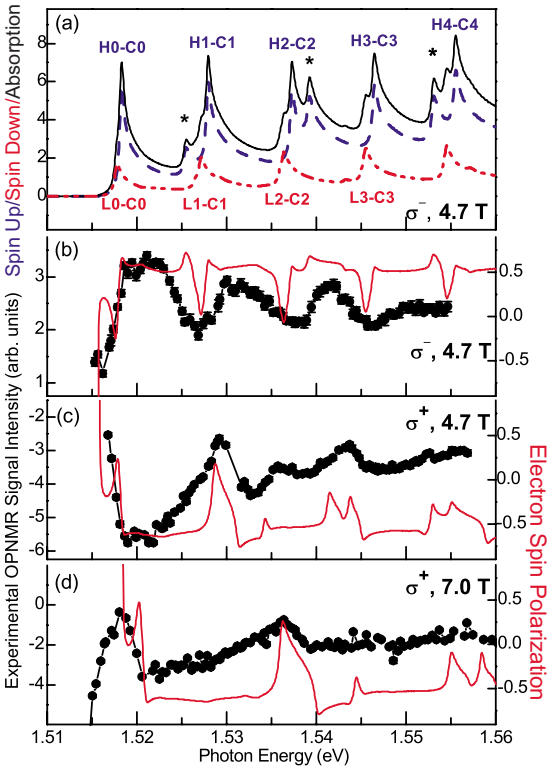

FIG. 3. (Color online) (a) Theoretical calculations of absorption of σ^- light by bulk GaAs at 4.7 T. Blue dashed line shows the absorption that produces spin-up electrons ($\alpha_\uparrow$, primarily from heavy hole transitions). Red dotted-dash line shows the absorption that produces spin-down electrons ($\alpha_\downarrow$, primarily from light-hole transitions). Black solid line shows total absorption, $\alpha_\uparrow+\alpha_\downarrow$. Peaks in (a) marked with an * result from valence-band mixing and are associated with heavy-hole spin-up to conduction-band spin-up transitions. Transitions between valence band heavy hole (H) or light hole (L) to conduction band (C) Landau levels are indicated by the labels in (a). (b)–(d) Depiction of the ^{69}Ga OPNMR signal intensity as a function of photon energy for: (b) σ^- polarized light and 4.7 T, (c) σ^+ polarized light and 4.7 T, and (d) σ^+ polarized light and 7.0 T. The experimental data (black symbols) are compared with the calculated electron polarization (solid red line), $(\alpha_\uparrow-\alpha_\downarrow)/(\alpha_\uparrow+\alpha_\downarrow)$.

The electrons recombine on a time scale much faster than the NMR time scale; therefore, the nuclear spins will experience an average electron spin polarization. Due to their fast spin-relaxation time, photoexcited holes do not contribute strongly to the OPNMR signal intensity.[27] Instead, the OPNMR of GaAs is dominated by the conduction-band spin polarization.

Results of our calculations for the spin-up absorption ($\alpha_\uparrow$), spin-down absorption ($\alpha_\downarrow$), and conduction-band polarization are shown in Fig. 3. In our simulations, we used σ^+ and σ^- circularly polarized light. In numerically calculating the integral in Eq. (1), the Dirac delta function in Fermi's golden rule is replaced by a Lorentzian. In Fig. 3(a), we plot the theoretical calculations of the total magnetoabsorption of

σ^- light (upper black line), spin-up absorption $\alpha_\uparrow$ (blue dashed line), and spin-down absorption $\alpha_\downarrow$ (red dotted-dashed line) in a magnetic field of 4.7 T. The theoretical calculations for all curves were shifted in energy by 6 meV to account for the shift due to Coulomb interactions (i.e., the exciton binding energy) which were not included in our calculations. The main transitions are labeled by the dominant character of the transition. For example, L0 to C0 is a light-hole Landau level 0 to conduction band Landau-level 0 transition. The "*" marks in Fig. 3(a) correspond to weaker transitions from heavy-hole Landau levels which result from valence-band mixing.

For σ^- excitation in Fig. 3(a), the total absorption (upper black line) is dominated by optical transitions from the heavy-hole spin-up Landau levels (solid black lines, Fig. 2) to conduction-band spin-up Landau levels. However, there are also optical transitions from the light-hole spin-up Landau levels (solid red lines, Fig. 2) to the conduction-band spin-down Landau levels for σ^- excitation. These light-hole transitions are difficult to see in absorption spectra since the light-hole transitions are weaker (by a factor of 3) than the heavy-hole transitions and are separated by only a few millielectron volts from the dominant heavy-hole transitions.

In Figs. 3(b)–3(d), we plot a combination of ^{69}Ga OPNMR experimental data (black symbols) at the two magnetic fields, 4.7 T and 7.0 T, for σ^- irradiation (at 4.7 T) and σ^+ irradiation (at 4.7 T and 7.0 T). (The OPNMR signal intensity at 7.0 T with σ^- irradiation is too weak to produce meaningful data; this attenuation of the OPNMR spectra is in agreement with theory.[9]) Superimposed onto the experimental OPNMR data are the calculated electron spin polarizations (red solid lines) for these fields and light helicities. It is evident that the oscillations of the OPNMR intensity are dependent on magnetic field, with peaks (and valleys) moving further apart as the external magnetic field strength is increased from 4.7 to 7.0 T. The oscillations in the OPNMR intensity follow a pattern that also depends on light helicity.

A plot of the electron spin polarization shows whether peaks in the absorption came from transitions from heavy- or light-hole Landau levels. When we look at the conduction-band spin polarization for σ^- excitation [red solid line, Fig. 3(b)], we see that the features which arise in the electron spin polarization are dominated by the transitions from the light-hole Landau levels. These light-hole spin-up to conduction-band spin-down transitions are very weak and barely visible in the plot of the total magnetoabsorption for σ^- excitation; however, these transitions are well resolved in the OPNMR data as a function of photon energy. We find that the conduction-band spin polarization is particularly sensitive to regions of photon energy where the total spin-polarized magnetoabsorption ($\alpha_\uparrow+\alpha_\downarrow$) and the differential magnetoabsorption ($\alpha_\uparrow-\alpha_\downarrow$) are different from one another, which occurs principally at the peaks of the light-hole transitions.

We see that the OPNMR experiments capture many of the features predicted by theory and are dominated by the light-hole-to-conduction-band transitions for optical pumping with both helicities of light. The experimental curves are much broader than the calculated curves, possibly due to broadening of the Landau levels from impurities in the sample.[17] Nevertheless, these weakly observed features produce demonstrable effects when examining the OPNMR spectra with respect to photon energy. By comparing shifts in the peak positions in the calculated magnetoabsorption (dominated by the stronger heavy-hole transitions) with the OPNMR signal—which is proportional to the conduction-band spin polarization (and dominated by the weaker light-hole transitions)—we can probe the spin splitting of the valence-band Landau levels shown in Fig. 2.

IV. CONCLUSIONS

In conclusion, we have demonstrated that oscillations observed with optical pumping above the band gap in OPNMR experiments can be correlated with detailed theoretical calculations of the Landau-level transitions from spin-split valence bands. The OPNMR oscillations are dominated by the weaker transitions from the light-hole Landau levels. Conventional optical magnetoabsorption primarily measures the threefold stronger transitions from the heavy-hole Landau levels while the OPNMR is inherently most sensitive to light-hole transitions, which generate large changes in the net electron spin polarization. Differences in the transition energies arise from spin splitting of the valence bands. We anticipate that OPNMR will become a powerful tool to investigate spin splitting of the electronic structures of both bulk and nanostructured semiconductors.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation under Grants No. DMR 0706313 and No. OISE 0530220, the National High Magnetic Field Laboratory, and a grant from the Office of Naval Research (ONR) under Contract No. 00075094. S.E.H. acknowledges support from the Alfred P. Sloan Foundation and the International Center for Advanced Renewable Energy and Sustainability (I-CARES) at WU.

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