Helicity independent optically-pumped nuclear magnetic resonance in gallium arsenide

Yunpu Li,1 Jonathan P. King,2 Le Peng,1 Maria C. Tamargo,3 Jeffrey A. Reimer,2 and Carlos A. Meriles1,4

1Department of Physics, City College of New York, CUNY, 138th Street and Convent Avenue, New York, New York 10031, USA
2Department of Chemical Engineering, University of California, 201 Gilman Hall, Berkeley, California 94720, USA
3Department of Chemistry, City College of New York, CUNY, 138th Street and Convent Avenue, New York, New York 10031, USA

(Received 15 November 2010; accepted 21 February 2011; published online 14 March 2011)

We present new phenomenology for optically-pumped nuclear magnetic resonance (OPNMR) of gallium arsenide. When pumping at low irradiation intensity, the OPNMR signal becomes independent of light helicity. The results are consistent with a mechanism in which the bulk signal represents competition between nuclear quadrupolar and electron-nuclear hyperfine relaxation. This mechanism is further supported by the scaling behavior of OPNMR for isotopes with varying hyperfine and quadrupolar interactions. These results indicate the magnitude and sign of nuclear polarization in the sample may be controlled as a function of depth by tuning photon energy and laser intensity, portending submicron scale patterning of nuclear magnetization. © 2011 American Institute of Physics. [doi:10.1063/1.3564897]

Optical control of nuclear polarization in semiconductors has been an active area of research for a community comprised of both nuclear magnetic resonance (NMR) spectroscopists and condensed matter physicists.1 As a probe of electron–nuclear interactions in the presence of spin-couplings, such control is interesting at a fundamental scientific level. There is also considerable interest in applying optical control of nuclear spin polarization to sensitivity-enhanced NMR (Ref. 2) and devices for quantum information processing.3,4 Control of the sign and magnitude of the polarization, especially as a function of position, is an important prerequisite for proposed devices.5 The most studied and best understood material is gallium arsenide (GaAs). Optically-pumped NMR (OPNMR) in GaAs has been explained by invoking a model where spin-polarized electrons are excited and spin exchange occurs with electrons bound at shallow donors.5 These bound, spin-polarized electrons then polarize proximate nuclei primarily through the Fermi-contact hyperfine interaction.

The energy dependence of the OPNMR signal amplitude in GaAs exhibits several general features. Far above the bandgap, the sign of the OPNMR signal may be controlled with the light helicity and the spectrum exhibits oscillations as a function of photon energy due to the appearance of Landau levels.9 Near the bandgap, a maximum in the OPNMR signal is observed due to the interplay of photon penetration depth and electron spin polarization.7 Below the bandgap, a different regime emerges where the OPNMR signal can no longer be controlled with light helicity and always assumes a positive value with respect to thermal polarization, suggesting a different polarization mechanism is at work (Fig. 1). Previous studies have proposed mechanisms, perhaps involving an electronic species with a positive g-factor10 but none have attempted to model this regime.

In this letter, we present experimental data showing the dependence of OPNMR signal on incident laser intensity, rather than photon energy. We identify a regime at low laser intensity with similar phenomenology to the previously observed regime of low photon energy. We show that both situations are explained by including the effects of both quadrupolar and electron-nuclear hyperfine relaxation on the growth and decay of the observed bulk OPNMR signal. The rates of both relaxation mechanisms are determined by the free electron concentration and shallow donor occupation fraction, which vary throughout the depth of the sample. These parameters are determined at a given laser intensity and photon energy by the optical absorption spectrum for GaAs.

OPNMR spectra of 71Ga, as well as 69Ga and 75As, were collected for a bulk semi-insulating GaAs wafer (American Institute of Physics, 2011). We present data on gallium arsenide, showing helicity-independent signal with positive NMR signals for low photon energy and for low laser intensity. Temperature is 8 K. 71Ga OPNMR spectra at different illumination intensities.
Crystal Technologies) 350 μm in thickness, surface orientation [100], resistivity greater than 107 Ω cm, mobility greater than 6000 cm2/V s, in a 9.4 T magnetic field at low temperature. The sample was placed in a continuous flow cryostat and in good thermal contact with a sapphire wafer to facilitate heat transfer at 6 to 8 K. NMR detection was preceded by the rf pulse sequence SAT\(−\)τ\(_L\)−τ\(_D\)−π/2, where τ\(_L\) and τ\(_D\) indicate illumination and dark intervals, which were 180 s and 10 s, respectively, unless otherwise noted. Saturation (SAT) prior to illumination was carried out via a series of π/2 pulses. The beam diameter was ~1.5 mm.

Figure 2 shows the dramatic change in observed NMR polarization for \(^{71}\)Ga when the laser intensity is decreased to ~0.83 μW/cm\(^2\) at 1.503 eV. At this photon energy, none of the previously published models\(^{11}\) can account for this pronounced loss of sensitivity to light helicity. A new mechanism for optically-induced NMR effects is therefore suggested. Previous workers have developed a model to explain the reduction in OPNMR signal by quadrupolar-induced depolarization.\(^{12}\) Here, we consider quadrupolar relaxation as a mechanism for polarization which, in certain regimes, may dominate over hyperfine polarization and reverse the sign of the NMR signal. Quadrupolar relaxation occurs near shallow donors as the capture, release, and recombination of electrons creates a fluctuating electric field gradient.\(^{12}\) These fluctuations are governed by the kinetic temperature of the electrons, and therefore, drive the nuclear spins to this temperature, with no dependence on the electron spin polarization. The equilibrium spin temperature achieved by quadrupolar relaxation results in a spin polarization which has the same sign as the thermal equilibrium spin polarization.

The important parameter in a model\(^{12}\) for OPNMR when nuclear spins possess a quadrupolar moment is the ratio of the two relaxation timescales, \(f\), defined by:

\[
f = \frac{T_{1,Q}}{T_{1,H}}.
\]  

where \(Q\) and \(H\) stand for quadrupolar and hyperfine relaxation. \(f\) is found to be a function of the donor occupation fraction, \(\Gamma\), and in the high magnetic field limit, this dependence is given by:

\[
f = f_0 \frac{\Gamma}{1 - \Gamma}.
\]  

Previous works have primarily studied the regime of high laser intensity and near to above-gap irradiation. In this case, \(\Gamma\) is close to one, \(f\) is large, and the hyperfine mechanism dominates. Here we consider the case where the donor occupation is less than one, either due to lower laser intensity or reduced absorption at low photon energies.

\(f_0\) is a property of the material and nucleus under investigation. By observing three different nuclei in the same sample, we can isolate the effects of varying the gyromagnetic ratio, \(\gamma\), and the product of the quadrupolar moment and electrostatic antishielding, \(QR^{14}\).\(^{12}\) \(f_0\) scales as:

\[
f_0 \propto \frac{\gamma^2}{Q^2R^{14}}.
\]  

Based on this scaling behavior, we expect the transition from quadrupolar-dominated to hyperfine-dominated behavior to occur at increasing values of laser intensity in the order \(^{71}\)Ga < \(^{69}\)Ga < \(^{75}\)As. This trend is clearly seen in Fig. 3, supporting the proposed mechanism.

It was discovered previously that laser penetration depth effects have a significant impact on the OPNMR signal.\(^{7,11}\) Our analysis predicts that not only can the magnitude of \(I_c\) vary throughout the depth of the sample but the sign of \(I_c\) may vary as well. Qualitatively, for high laser intensity and near to above gap irradiation, the majority of the light is absorbed near the surface of the sample, with a high donor occupation fraction and free electron concentration. The OPNMR signal is then almost entirely due to this region where hyperfine relaxation is dominant. As the photon energy is lowered well below the bandgap, the penetration depth becomes large, the rate of absorption per unit volume is much lower and is spread throughout a larger region of the sample. In this case, the OPNMR signal is due to a large region of the sample with lower donor occupation fraction and free electron concentration where quadrupolar relaxation dominates. This accounts for the behavior in the low photon energy regime that was previously unexplained.\(^{10}\)

For near and above gap irradiation, the OPNMR signal will always be limited to the region near the surface due to small penetration depths. By reducing the laser intensity, however, the free electron concentration and donor occupa-
tion fraction in this region may be lowered so that quadrupolar relaxation dominates. There also exists an intermediate case for both laser intensity and photon energy where the net OPNMR signal is zero. This analysis suggests that in this case \( \langle I_z \rangle \) is not zero throughout the sample but rather there are regions of positive and negative \( \langle I_z \rangle \).

We have experimentally demonstrated a new, helicity-independent regime of OPNMR at low laser intensity. These data are consistent with competing hyperfine and quadrupolar relaxation mechanisms, including scaling behavior for different nuclei. This physical picture also accounts for the previously unexplained regime of low photon energy. These competing mechanisms provide a means by which to pattern nuclear polarization on a submicron length scale by adjusting photon energy and laser intensity. This patterning is an important step in the development of devices using hyperpolarized nuclear spins.

J.P.K. acknowledges funding from the NDSEG Program, C.A.M. acknowledges support from the Research Corporation, and all the authors acknowledge support from the National Science Foundation under Project No. ECS-0608763. Y. Li and J. P. King contributed equally to this work.