# Nuclear spin-lattice relaxation in *n*-type insulating and metallic GaAs single crystals

J. Lu, M. J. R. Hoch, P. L. Kuhns, W. G. Moulton, Z. Gan, and A. P. Reyes

National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310, USA

(Received 15 May 2006; revised manuscript received 27 July 2006; published 29 September 2006)

The coupling of electron and nuclear spins in *n*-GaAs changes significantly as the donor concentration *n* increases through the insulator-metal critical concentration  $n_{\rm C} \sim 1.2 \times 10^{16}$  cm<sup>-3</sup>. The present measurements of the <sup>71</sup>Ga relaxation rates *W* made as a function of magnetic field (1–13 T) and temperature (1.5–300 K) for semi-insulating GaAs and for three doped *n*-GaAs samples with donor concentrations  $n=5.9 \times 10^{15}$ ,  $7 \times 10^{16}$ , and  $2 \times 10^{18}$  cm<sup>-3</sup>, show marked changes in the relaxation behavior with *n*. Korringa-like relaxation is found in both metallic samples for T < 30 K, while for T > 30 K phonon-induced nuclear quadrupolar relaxation is dominant. The relaxation rate measurements permit determination of the electron probability density at <sup>71</sup>Ga sites. A small Knight shift of -3.3 ppm was measured on the most metallic ( $2 \times 10^{18}$  cm<sup>-3</sup>) sample using magic-angle spinning at room temperature. For the  $n=5.9 \times 10^{15}$  cm<sup>-3</sup> sample, a nuclear relaxation model involving the Fermi contact hyperfine interaction, rapid spin diffusion, and exchange coupled local moments is proposed. While the relaxation rate behavior with temperature for the weakly metallic sample,  $n=7 \times 10^{16}$  cm<sup>-3</sup>, is similar to that found for the just-insulating sample, the magnetic field dependence is quite different. For the  $5.9 \times 10^{15}$  cm<sup>-3</sup> sample this results in an increase in the relaxation rate ascribed to an increase in the density of states at the Fermi level as the Landau level degeneracy is increased.

DOI: 10.1103/PhysRevB.74.125208

PACS number(s): 71.30.+h, 76.60.-k, 82.56.Na

### I. INTRODUCTION

Semiconductors undergo a metal-insulator transition (MIT) at low temperature for a critical dopant concentration  $n_{\rm C}$ . There has been continuing interest in studying the MIT in semiconductors, such as Si:P, using a variety of techniques including NMR. However, to our knowledge, no systematic NMR investigation of the technologically important direct gap semiconductor *n*-GaAs has been carried out across the MIT. The present work involves low *T* nuclear relaxation rate measurements on samples with donor concentrations above and below the transition. The Knight shift has been measured in the most metallic sample.

The electron-nucleus hyperfine interaction in GaAs is important in developing GaAs based electron spin devices.<sup>1</sup> Dynamic nuclear polarization (DNP) experiments in GaAs have shown the importance of hyperfine interactions in these processes.<sup>2-5</sup> Recent optically pumped DNP experiments in GaAs have been interpreted as showing that the coupling between itinerant electrons and host lattice nuclei is important in the DNP mechanism;<sup>5</sup> this is in contrast to an earlier model involving localized donor states exchange-coupled to carriers excited into the conduction band from the valence band.<sup>2-4</sup> The hyperfine interaction plays an essential role in the recently reported dc current-induced DNP in GaAs.<sup>6,7</sup> The present <sup>71</sup>Ga relaxation rate measurements on a set of GaAs samples with donor concentrations spanning the MIT provide information on the hyperfine interaction as the electronic structure evolves from localized donor states to itinerant carriers.

McNeil and Clark have measured the nuclear relaxation rate of undoped GaAs from 4 K to room temperature.<sup>8</sup> Their results show that at temperatures above 30 K phononmediated quadrupolar relaxation is dominant and this mechanism may be expected to determine relaxation rates in the high temperature range for all donor concentrations. The present work has therefore concentrated on the relaxation properties of n-GaAs at low temperatures where electron-nucleus interactions are important.

# **II. EXPERIMENT**

The semi-insulating and *n*-doped GaAs used in the experiments are referred to as follows: (1) SI (semi-insulating), (2)  $5.9 \times 10^{15}$  ( $n=5.9 \times 10^{15}$  cm<sup>-3</sup>), (3)  $7 \times 10^{16}$  ( $n=7 \times 10^{16}$  cm<sup>-3</sup>), and (4)  $2 \times 10^{18}$  ( $n=2 \times 10^{18}$  cm<sup>-3</sup>). The samples were obtained from American Crystal Technology Inc. (samples 1 and 4) and the Institute of Electronic Materials Technology, Warsaw (samples 2 and 3). The compensation ratios for the samples have not been determined.

For the NMR experiments, small pieces were cleaved from (001) GaAs wafers, giving samples with dimensions  $\sim 3 \times 4 \times 0.4$  mm<sup>3</sup>, and the applied magnetic field was aligned along one cleaved (110) direction with the axis of the rf coil along the orthogonal (110) direction. Temperatures in the range 1.4 K-300 K were obtained using a pumped He cryostat and a Lakeshore temperature controller. Nuclear relaxation rate measurements were made using a computercontrolled coherent pulsed NMR spectrometer and a variable field (corrected but unshimmed) superconducting magnet with 10 ppm/mm DSV homogeneity. A powder sample of <sup>27</sup>Al loaded together with the GaAs samples was used for determining optimal pulse length conditions. Relaxation rates  $W=1/T_1$  (with  $T_1$  the spin-lattice relaxation time) were measured by saturating the nuclear magnetization using a comb of 90° pulses, and monitoring the free-induction decay (FID) recovery signal for 16 different delay times up to 5  $T_1$ . The signal-to-noise ratio for the equilibrium signal at 4.2 K is  $\sim$ 2000. The line shape is approximately Lorentzian with a width of  $\sim 80$  ppm due to the combined effects of dipolar



FIG. 1. (Color online) Electrical resistivities of the  $5.9 \times 10^{15}$  and  $7 \times 10^{16}$  GaAs samples as function of (a) 1/T and (b) *H*.

broadening and magnet inhomogeneity over the sample length. All FID recovery curves were well fit with a single exponential function using a least squares fitting procedure. The statistical error obtained from the  $\chi^2$  values is typically less than 1%, but to allow for systematic errors such as instrumental drifts we allow an error of 10% in our measured *W* values. Spin-echo methods were used to determine  $T_2$  for <sup>71</sup>Ga.

For the resistivity measurements, samples of comparable size to the NMR samples were prepared with two Ohmic contacts made using indium followed by annealing at 410 °C in forming gas for 10 min. The quality of the Ohmic contacts was checked using room temperature resistivity measurements to ensure that the measured values agreed with the nominal values.

## **III. RESULTS AND DISCUSSION**

The resistivities of samples  $5.9 \times 10^{15}$  and  $7 \times 10^{16}$  in H = 0 are shown versus 1/T in Fig. 1(a). For sample 5.9  $\times 10^{15}$ , the resistivity changes exponentially with 1/T for T > 10 K consistent with an increase with T of the number of conduction electrons, due to thermal activation of localized donors. The ln  $\rho$  versus 1/T data give a straight line of slope  $\Delta \sim 2$  meV, in agreement with Ref. 9. At temperatures below 10 K, the resistivity data of sample  $5.9 \times 10^{15}$  can be fit with  $\ln(\rho) \sim T^{1/2}$  consistent with variable range hopping conduction.<sup>10</sup> For the just-metallic sample  $7 \times 10^{16}$ , the resistivity consistent with variable range hopping conduction.



FIG. 2. (Color online) (a) Log-log plot of <sup>71</sup>Ga W as function of T for the insulating and metallic GaAs samples in H=1.553 T. (b) Linear plot of W vs T in the low-temperature region showing Korringa-type relaxation for the two metallic samples. The solid lines are linear fits for the metallic samples, and the dashed line is a  $\sim T^{0.4}$  fit for the 5.9 × 10<sup>15</sup> sample.

tivity is independent of temperature below 20 K. Above 20 K, the resistivity decreases with increasing temperature due to the changes in mobility.<sup>11</sup>

The dependence of the resistivity at 4.2 K on magnetic field in the range 1–15 T is shown in Fig. 1(b). The large positive magnetoresistance found in sample  $5.9 \times 10^{15}$  is the well-known magnetic freeze-out effect<sup>9</sup> due to the shrinking of the donor electron wave function (Bohr radius) in high-magnetic fields. The positive magnetoresistance in sample  $7 \times 10^{16}$  is much smaller than that of sample  $5.9 \times 10^{15}$  but increases significantly for H>8 T. This effect is linked to the decrease in the magnetic length  $a_H = (\hbar/eH)^{1/2}$  to values less than the average donor separation distance.<sup>12</sup>

Figure 2(a) shows the <sup>71</sup>Ga nuclear spin-lattice relaxation rate W as a function of T for all samples. The lines in the figure are guides to the eye. At high T the relaxation curves converge consistent with the behavior of undoped GaAs in Ref. 8, where quadrupolar relaxation is dominant at temperatures above  $\sim$ 30 K. The W data of sample SI drop off rapidly with temperature and below 30 K the relaxation rate is at least one order of magnitude smaller than that of the other samples. This suggests that below 30 K the spin-lattice re-

laxation is controlled by donors. At higher temperatures, comparable to the Debye temperature of 360 K,<sup>13</sup> W increases approximately as  $T^2$ , a signature of two phonon quadrupolar relaxation. This is consistent with the results of Ref. 8. Figure 2(b) shows W versus T for the low-temperature region. Sample  $2 \times 10^{18}$  shows linear Korringa dependence of the relaxation rate on T as expected for a metallic sample, while sample  $7 \times 10^{16}$  shows approximately Korringa behavior over the low-temperature range. Despite a factor of 12 difference in doping concentration and the large difference in low-T resistivity between samples  $5.9 \times 10^{15}$  and  $7 \times 10^{16}$ , the difference in their relaxation rate at T < 10 K is subtle. This suggests that the spin relaxation by localized donors in the insulating sample is  $\sim 10$  times more efficient than that of itinerant carriers in the metallic sample. Relaxation on the insulating side of the MIT is attributed to the interaction between localized electrons and nuclear spins in combination with rapid spin diffusion, as discussed in detail below.

For metallic samples with  $n > n_{\rm C}$ , spin relaxation is due to scattering processes involving conduction electrons close to the Fermi level. When the temperature is well below the Fermi temperature  $T_{\rm F}$ , W is proportional to T and in the single-electron approximation is given by<sup>14</sup>

$$W = (64/9) \pi^{3} \gamma_{e}^{2} \gamma_{n}^{2} \hbar^{3} |\phi_{\rm F}(0)|^{4} k_{\rm B} T \rho^{2}(E_{\rm F})$$
  
=  $4 \pi^{3} \gamma_{e}^{2} \gamma_{n}^{2} \hbar^{3} \Omega^{2} n^{2} P_{f}^{2} T / (k_{\rm B} T_{F}^{2}),$  (1)

with  $\gamma_e$  and  $\gamma_n$  the gyromagnetic ratios for the free electron and the nucleus respectively,  $\hbar$  Planck's constant,  $\rho(E_{\rm F})$  the density of states at the Fermi level  $E_{\rm F}$  and  $k_{\rm B}$  Boltzmann's constant.  $\phi_{\rm F}(0)$  is the wave function of conduction electrons at a nuclear site, n is the donor concentration.  $P_f$  $=\Omega^{-1}|\phi_{\rm F}(0)|^2$  is the electron probability density at the nucleus normalized in the unit cell volume  $\Omega$ . In the free electron gas model,  $T_{\rm F}$  is proportional to  $n^{2/3}$ , and from Eq. (1) it follows that W is proportional to  $n^{2/3}$ . This leads to the prediction that  $T_1$  of sample  $7 \times 10^{16}$  should be about nine times longer than that of sample  $2 \times 10^{18}$ , which is confirmed by our experimental results. Similar nuclear relaxation results have been obtained in the metallic phase of Si:P.15 Using the effective electron mass approximation  $(m_e)$ =0.067 $m_{\rm o}$ ), the *n*-GaAs Fermi temperatures  $T_{\rm F}$  are estimated to be about 110 K and 1000 K for samples  $7 \times 10^{16}$  and 2  $\times 10^{18}$ , respectively. With these values and our W data, we use Eq. (1) to estimate  $P_f$  from our experimental relaxation times and obtain <sup>71</sup>Ga  $P_f = 7 \times 10^{25}$  cm<sup>-3</sup> and  $8 \times 10^{25}$  cm<sup>-3</sup> for samples  $2 \times 10^{18}$  and  $7 \times 10^{16}$  respectively. (We note that the single-particle parabolic band approximation may not be adequate for the weakly metallic sample  $7 \times 10^{16}$ .) The estimated  $P_f$  values are comparable with that calculated for atomic <sup>71</sup>Ga ( $6.8 \times 10^{25}$  cm<sup>-3</sup>, Ref. 16) as well as the previous estimates ( $P_f = 5.8 \times 10^{25}$  cm<sup>-3</sup>, Ref. 2) made for the <sup>71</sup>Ga site in optically pumped dynamic polarization experiments on GaAs. It is clear that the Korringa mechanism accounts for the low-T, low-H nuclear relaxation behavior for both metallic GaAs samples. In higher fields allowance must be made for the evolution of the Landau levels in the plane perpendicular to the field resulting in modifications to Eq. (1).<sup>17</sup> Changes in the density of states at  $E_{\rm F}$  due to the Landau levels are introduced in connection with the *W* versus *H* results discussed below.

The Knight shift K for  $^{71}$ Ga in GaAs can be predicted using our low temperature W values in the Korringa relation,<sup>18</sup>  $K^2T/W = (g^* \mu_B)^2/(4\pi k_B \hbar \gamma_n^2)$ , where  $g^* = -0.44$  is the effective g factor for GaAs. On this basis |K| is calculated to be 11 ppm for the most metallic sample  $2 \times 10^{18}$ . The linewidth of  $\sim 80$  ppm prevented us from measuring the small Knight shift with our low-temperature NMR spectrometer. A room temperature 10 kHz magic-angle spinning probe in a 19.6 T high-resolution superconducting magnet was therefore used to measure K for a powder sample of the most metallic wafer  $(2 \times 10^{18})$  with powdered SI as a reference and this gave  $K \sim -3.3$  ppm. Allowance for temperature effects can be made using the expression<sup>15</sup>  $K = \pi \gamma_e^2 \hbar^2 (P_f / k_B T_F) [1 - \pi^2 / 12 (T/T_F)^2]$  but with  $T_F \sim 1000$  K for the  $2 \times 10^{18}$  sample, the correction is small, approximately 7.5% at 300 K. The discrepancy between the Korringa relation predicted 11 ppm and the measured value at 300 K in 19.6 T is not understood at present. Further low-T, low-H Knight shift measurements are needed to resolve this issue.

For sample  $5.9 \times 10^{15}$ , the transport data in Fig. 1 show that below 10 K the donor electrons are highly localized. The acceptor centers, having captured an electron from a donor center, are not paramagnetic and do not contribute to nuclear relaxation. However, other deep level paramagnetic impurities may contribute to relaxation. Measurements of W versus T for the SI sample shown in Fig. 2 indicate that such contributions are roughly an order of magnitude smaller than that of the localized donors in sample  $5.9 \times 10^{15}$ . Assuming doping does not introduce a significant amount of other impurities, we conclude that the relaxation by other paramagnetic impurities is not important in our analysis.

For insulating GaAs with highly localized electrons, the low-T nuclear relaxation process is likely to involve spin diffusion. Spin diffusion in solids containing paramagnetic impurities has been extensively studied as a mechanism for relaxing nuclear spins via paramagnetic impurities. The spin diffusion equation has been solved for various cases.<sup>19-22</sup> When spin diffusion is rapid, the relaxation rate is obtained as a spatial integral of direct relaxation over the region in which spin diffusion operates.<sup>20</sup> We show below that this case applies to just-insulating GaAs. In the usual treatment of nuclear relaxation by paramagnetic impurities, it is assumed that the dipolar interaction is the dominant direct relaxation mechanism.<sup>14,19-22</sup> This assumption is valid when the impurity wave function is localized in a region very small compared with the distance between impurities. However, for GaAs where the electron Bohr radius is large, the Fermi contact interaction is much stronger than dipolar interaction wherever the electron wave function is not negligible,<sup>23</sup> and we demonstrate below that for insulating GaAs where the Bohr radius of the shallow donors  $a_0$  $\sim 100$  Å,<sup>2-4</sup> the Fermi contact hyperfine is dominant when the distance between impurities is less than 500 Å.

We now show that the spin diffusion is rapid compared with the spin-lattice relaxation process. The spin diffusion coefficient D for a particular nuclear species can be estimated using<sup>14</sup>



FIG. 3. (Color online) Cartoon depicting the relaxation model used for insulating GaAs. Inside the diffusion barrier radius *b* the hyperfine field is sufficiently large to block spin diffusion. For r > b, spin diffusion is fast and establishes a spin temperature for spins in this region while r < c is the "wipe-out" region where the nuclei experience large shifts and only make a contribution to the tails of the NMR spectrum. The outer radius *d* is taken as half the average spacing between donor sites.

$$D \sim Wa^2 \sim (\sqrt{M_2}/30)a^2,$$
 (2)

where *a* is the distance between nearest-neighbor <sup>71</sup>Ga nuclei, the factor *W* is the probability of a mutual spin flip of these two nuclei with  $M_2$  the dipolar second moment for like spins. We have calculated  $M_2$  for <sup>71</sup>Ga in GaAs and obtain an average  $D \sim 2.0 \times 10^{-13}$  cm<sup>2</sup>/s. (Measurement of the <sup>71</sup>Ga spin-spin relaxation time  $T_2$  using spin-echo methods gives  $T_2 \sim 400 \ \mu s$  and using  $D \sim a^2/50T_2$  (Ref. 19) gives a value consistent with the above estimate.) In estimating the spin diffusion time  $t=d^2/D$ , we take the distance *d* as half the average distance between donor sites, or  $d=0.5/n^{1/3} \sim 270$  Å, and obtain  $t \sim 36$  s. The measured  $T_1$  of sample  $5.9 \times 10^{15}$  at low temperatures is roughly two orders of magnitude longer than this. Therefore, spin diffusion is not the limiting process, or bottleneck, in nuclear spin relaxation and the rapid diffusion case applies in this sample.

A cartoon sketch showing the essentials of the model presented here for the  $5.9 \times 10^{15}$  sample is shown in Fig. 3. Inside the diffusion barrier radius b, the large local field suppresses spin diffusion since neighboring like spins see different average fields that produce shifts in the resonance frequency larger than the linewidth. Outside this sphere, spin diffusion becomes of dominant importance with the outer sphere radius d roughly half the average distance between donors. (In this random system the geometrical details will vary from donor site to donor site.) Some spins inside the diffusion barrier, but outside what is termed the wipe-out radius c < b, within which nuclei experience shifts several times larger than the linewidth, will make a contribution in the tails of the NMR spectrum. Such contributions to the signal will be small because the volume associated with the shell of spins between c and b compared to the volume of the shell between radii b and d is small in insulating GaAs.

In general, the electron-nuclear interaction can be written  $as^{14}$ 



FIG. 4. (Color online) Plot of the static hyperfine fields for the contact hyperfine interaction  $H_{hf}$  and the dipolar interaction  $H_{dip}$  as a function of distance *r* from a donor site. The hyperfine field has been scaled by a factor  $10^{-3}$ .

$$\mathcal{H} = \mathcal{H}_{\rm hf} + \mathcal{H}_{\rm dip} = (16\pi/3)\gamma_n \hbar \mu_{\rm B} (\boldsymbol{I} \cdot \boldsymbol{S}) |\varphi(\boldsymbol{R})|^2 + 2\mu_{\rm B}\gamma_n \hbar \boldsymbol{I} \cdot \operatorname{grad}_{\rm R} \int \{\operatorname{div}[\boldsymbol{S}\rho(\boldsymbol{r})]/|\boldsymbol{r} - \boldsymbol{R}|\} d^3r, \quad (3)$$

where  $\mathcal{H}_{hf}$  and  $\mathcal{H}_{dip}$  represent the Fermi contact and dipolar interactions, respectively. We use the envelope function  $F(r) = (1/\pi a_0^{-3})^{1/2} \exp(-r/a_0)$  to obtain  $\rho(r) = |F(r)|^2/(1/\pi a_0^{-3}) \exp(-2r/a_0)$ , where  $a_0$  is the Bohr radius,<sup>24</sup> while for the Fermi contact term,  $|\varphi(\mathbf{R})|^2 = \Omega P_f |F(r)|^2$ . We take  $a_0 = 100$  Å,  $P_f = 7 \times 10^{25}$  cm<sup>-3</sup> in evaluating both terms in Eq. (3) numerically along the electron spin quantization direction *z*. The results are plotted in Fig. 4 in gauss. (Note that in the plot the hyperfine field is reduced by a factor of  $10^3$ .) It is evident that for r < 300 Å, the Fermi contact interaction along *z* is much larger than the dipolar interaction. Similar results to within an order of magnitude are predicted along other directions. Based on this analysis, we neglect the dipolar interaction in the following discussion.

In the case of rapid spin diffusion, analogous to the treatment by Blumberg for the dipolar relaxation case,<sup>20</sup> the relaxation rate via the fluctuating hyperfine interaction can be expressed as

$$W = 4\pi n \int_{b}^{d} K(r) r^{2} dr, \qquad (4)$$

where  $K(r) \sim CJ(\omega)P_f^2 \Omega^2 a_o^{-6} \exp(-4r/a_o)$ ,  $C = [(16\pi/3)\gamma_n g\mu_B]^2 S(S+1)$ , and *d* is half of the average distance between donors. *b* is the diffusion barrier radius defined as the distance from a donor site at which the average local field seen by nuclei is equal to the linewidth. For convenience we take the upper limit  $d = \infty$ , because K(r) at *d* is very small and the integral converges.  $J(\omega)$  with  $\omega = \omega_e$   $-\omega_n$  is the spectral density. For simplicity, we choose the Debye form of the spectral density  $J(\omega) = \tau/[1+(\omega_e -\omega_n)^2\tau^2]$ , where  $\omega_e$  and  $\omega_n$  are the resonance frequencies for the electron and nucleus, respectively, and  $\tau$  is the electron correlation time arising from electron-electron interaction or thermally induced process. Other forms for  $J(\omega)$  have been proposed allowing for the distribution of  $\tau$  values but they involve additional parameters.<sup>26</sup>

Integration of Eq. (4) gives

$$W = 4\pi n C J(\omega) P_f^2 \Omega^2 a_0^{-6} (b^2 + b a_0 / 2 + a_0^2 / 8)$$
  
×(a\_0 / 4) exp(-4b/a\_0). (5)

It is evident that W varies with temperature and magnetic field via  $J(\omega)$ , b, and  $a_0$ . In order to make numerical estimates of the relaxation rate, it is necessary to consider the electron correlation time  $\tau$  and the diffusion barrier radius b, the distance from a donor site at which the average field produced by the electron moment is equal to the linewidth. We first estimate  $\tau$ . The fluctuations of the Fermi contact hyperfine field at nuclear sites can come from two mechanisms. The first mechanism involves fluctuations in donor site occupancy either through thermal activation from the band to the conduction band or via impurity variable-range-hopping.<sup>25</sup> The latter process is responsible for electrical conduction at low temperatures as indicated by the resistivity versus T curve in Fig. 1(a). For the sample  $5.9 \times 10^{15}$ , we find the data can be fitted by  $W \propto T^{0.4}$  below 10 K [Fig. 2(b)]. This weak T dependence points away from thermal excitation processes being involved in spin-lattice coupling in the low-T region. A second possible mechanism involves the magnetic exchange coupling between localized donors.<sup>26</sup> This coupling is thought to determine the long electron spin lifetime in lightly doped GaAs.<sup>27,28</sup> The exchange coupling of two localized donors can be written as<sup>28</sup>

$$J(R) = -0.82(2E_{\rm B})(R/a_{\rm o})^{5/2}\exp(-2R/a_{\rm o}), \qquad (6)$$

where  $E_{\rm B}$  is the localized electron binding energy (~6 meV) and *R* is the distance between the two donors. The correlation time for the exchange process is given by  $\tau_{\rm c} \sim \hbar/J(R)$ . For sample  $5.9 \times 10^{15}$ , assuming a Poisson distribution, the most *probable* distance between neighboring donors is R=450 Å. Taking  $a_0=100$  Å,  $\tau$  is calculated to be ~2.5 × 10<sup>-11</sup> s. Clearly there is a distribution of correlation times,<sup>26</sup> corresponding to a distribution of *R* values, but we shall assume that this estimate of  $\tau$  is sufficiently reliable for our purposes. Since J(R) is temperature independent, so is  $\tau$  in the exchange model.

We next estimate the diffusion barrier b. Neglecting the dipolar term in Eq. (3), the average field at a nuclear site at distance r from an occupied donor site is given by

$$H_e(\mathbf{r}) = (16\pi/3)g\mu_{\rm B}\langle \mathbf{S} \rangle |\varphi(\mathbf{r})|^2, \qquad (7)$$

where  $\langle S \rangle$  is the time average of electron spin *S*, and for short correlation times compared to nuclear  $T_2$ , such as in our case,  $\langle S \rangle = 1/2 \tanh(g\mu_B H/k_B T)$ . At the diffusion barrier radius r = b, we take  $H_e(b)$  to be of the order of  $\Delta H$ , where  $\Delta H$  is the nuclear linewidth. We obtain the linewidth based on our calculated second moment of <sup>71</sup>Ga spins as  $\Delta H \sim 0.5$  G. Assuming donor occupancy close to 1, we find *b* is temperature and field dependent and is on order of  $\sim 200$  Å, comparable with the value of 140 Å obtained by Paget<sup>3</sup> for <sup>75</sup>As in GaAs at 1.7 K in a 0.6 T field. Using our estimates of  $\tau$  and *b* together with the other numbers given above, we have cal-



FIG. 5. <sup>71</sup>Ga W in GaAs as function of field H for three different doping levels at 4.2 K. The just-insulating sample  $5.9 \times 10^{15}$  and the weakly metallic sample  $7 \times 10^{16}$  show quite different behavior with increasing H as discussed in the text.

culated W from Eq. (5) for H=1.55 T, T=4 K obtaining a value of  $\sim 6.2 \times 10^{-4}$  s<sup>-1</sup> in fair agreement with the experimental results of  $8 \times 10^{-4}$  s<sup>-1</sup>. This finding provides support for the proposed relaxation model involving antiferromagnetic exchange coupled localized electron spins relaxing nuclei through hyperfine coupling in the fast diffusion limit.

Following Eq. (5), a simple analysis shows that an increase in *T* between 1 and 30 K at 1.55 T leads to a decrease in *b* and consequently a near linear increase in *W*. In our analysis we use the contact hyperfine interaction, and find that the predicted *T* dependence is similar to that obtained by Blumberg for the dipolar *I*-*S* coupled rapid diffusion case in the short correlation time limit.<sup>20</sup> A linear *T* dependence for *W* is not experimentally observed. The experimental results show  $\sim T^{0.4}$  dependence rather than a linear *T* dependence. It is likely that for T > 10 K, thermal activation of donor electrons leads to changes in  $J(\omega)$  by changing  $\tau$ . Further low-temperature experiments are needed to better test the *T* dependence of our model.

Turning to the *H* dependence, our analysis based on Eq. (5) predicts that increasing *H* at 4.2 K will lead to a slow increase in *b* and a corresponding decrease in *W*. The analysis is in qualitative agreement with our data for the 5.9  $\times 10^{15}$  sample shown in Fig. 5. However, for H > 6 T, if we replace  $a_0$  with  $a_H = (\hbar/eH)^{1/2}$ , the magnetic length, to account for the shrinkage in  $a_0$  in high fields, the predicted  $W \sim H$  dependence is much stronger than found in the experiments. This suggests that more elaborate calculations are required in this case since the electron wave function is no longer approximately spherical.

Figure 5 also shows the *W* magnetic field dependence for metallic samples  $7 \times 10^{16}$  and  $2 \times 10^{18}$  measured at 4.2 K. The significant increase of *W* with *H* in the weakly metallic sample  $7 \times 10^{16}$  can be explained, making use of Eq. (1), in terms of an increase in the density of states at high field (*H*>4 T). For sample  $7 \times 10^{16}$ , following Ref. 17, it is estimated that all electrons are in the first Landau level when *H*>4 T. With increasing *H*, the density of states increases due to the increase in Landau level degeneracy. We did not observe quantum oscillations in lower fields where the higher Landau levels are filled. This may be due to Landau level broadening as a result of doping-induced disorder in this sample.<sup>12</sup> We note that *W* appears to saturate at the highest field of 13 T. Further investigation of this field effect is needed in just-metallic samples. No significant change in *W* with *H* is observed in the highly metallic sample  $2 \times 10^{18}$ .

# **IV. CONCLUSION**

Measurements of the spin lattice relaxation rate W of <sup>71</sup>Ga in *n*-GaAs as function of temperature and magnetic field for samples doped both below and above the MIT have been carried out. For all samples at temperatures above 30 K, quadrupolar relaxation through phonon scattering is the dominant process. At temperatures below 30 K, Korringa relaxation behavior is observed for the two metallic samples. The parabolic band single-electron model involving scattering of electrons close to the Fermi level is used to interpret these results. It is suggested that the increase in W with magnetic field in the sample doped just above the MIT is linked to changes in the density of states at the Fermi energy in the first Landau level with increasing H. Knight shift predictions made using the Korringa relation (11 ppm) are compared

with experimental values (-3.3 ppm) obtained at room temperature and in high field. The discrepancy in the values is much larger than estimated uncertainties. Further high-resolution experiments at low temperatures and in low fields are needed to resolve the matter.

The doped insulating sample  $(n/n_{\rm C} \sim 0.5)$  shows a weak temperature dependence of W for T < 30 K. The relaxation model proposed for this case involves the Fermi contact hyperfine interaction and rapid nuclear spin diffusion. AF exchange interactions among localized donors produce fluctuating hyperfine fields at nuclear sites and a low-T average correlation time  $\tau_{\rm c}$  of  $\sim 2.5 \times 10^{-11}$  s is estimated. Our model qualitatively explains the decrease in W both with decreasing T and with increasing H in this sample.

### ACKNOWLEDGMENTS

This work is financially supported by the Defense Advanced Research Projects Agency (DARPA) Grant No. MDA972-03-01-0010. Partial support by the National Science Foundation under Cooperative Agreement No. DMR-0084173 and the State of Florida is gratefully acknowledged.

- <sup>1</sup>I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 1 (2004).
- <sup>2</sup>D. Paget, G. Lampel, B. Sapoval, and V. I. Safarov, Phys. Rev. B **15**, 5780 (1977).
- <sup>3</sup>D. Paget, Phys. Rev. B **25**, 4444 (1982).
- <sup>4</sup>P. L. Kuhns, A. Kleinhammes, T. Schmiedel, W. G. Moulton, E. Hughes, S. Sloan, P. Chabrier, and C. R. Bowers, Phys. Rev. B 55, 7824 (1997).
- <sup>5</sup>A. K. Paravastu, S. E. Hayes, B. Schwickert, L. N. Dinh, M. Balooch, and J. A Reimer, Phys. Rev. B **69**, 075203 (2004); A. K. Paravastu and J. A. Reimer, Phys. Rev. B **71**, 045215 (2005).
- <sup>6</sup>M. J. R. Hoch, J. Lu, P. L. Kuhns, W. G. Moulton, and A. P. Reyes, Phys. Rev. B **72**, 233204 (2005).
- <sup>7</sup>A. K. Paravastu, P. J. Coles, J. A. Reimer, T. D. Ladd, and R. S. Maxwell, Appl. Phys. Lett. **87**, 232109 (2005).
- <sup>8</sup>J. A. McNeil and W. G. Clark, Phys. Rev. B 13, 4705 (1976).
- <sup>9</sup>T. O. Poehler, Phys. Rev. B **4**, 1223 (1971).
- <sup>10</sup>N. F. Mott, *Metal-insulator Transitions* (Taylor and Francis, New York, 1990).
- <sup>11</sup>V. C. Kieu, Phys. Status Solidi A 45, 571 (1978).
- <sup>12</sup> M. I. Dyakonov, A. L. Efros, and D. L. Mitchell, Phys. Rev. 180, 813 (1969).

- <sup>13</sup>J. S. Blakemore, J. Appl. Phys. **53**, R123 (1982).
- <sup>14</sup>A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, 1961).
- <sup>15</sup>E. M. Meintjes, J. Danielson, and W. W. Warren, Jr., Phys. Rev. B **71**, 035114 (2005); M. J. R. Hoch and D. F. Holcomb, Phys. Rev. B **71**, 035115 (2005).
- <sup>16</sup>J. Morton and K. F. Preston, J. Magn. Res. **30**, 577 (1978).
- <sup>17</sup>F. Bridges and W. G. Clark, Phys. Rev. **182**, 463 (1969).
- <sup>18</sup>P. Braun and S. Grande, Phys. Status Solidi B 74, 209 (1976).
- <sup>19</sup>N. Bloembergen, Physica (Amsterdam) **15**, 386 (1949).
- <sup>20</sup>W. E. Blumberg, Phys. Rev. **119**, 79 (1960).
- <sup>21</sup>I. J. Lowe and D. Tse, Phys. Rev. 166, 279 (1968).
- <sup>22</sup>G. R. Khutsishvili, Sov. Phys. Usp. **11**, 802 (1969).
- <sup>23</sup>C. Deng and X. Hu, Phys. Rev. B **72**, 165333 (2005).
- <sup>24</sup>W. Kohn, Solid State Physics Vol. 5, 257–320 (Academic Press, New York, 1957).
- <sup>25</sup>B. I. Shklovskii and A. L. Efros, *Electronic Properties of Doped Semiconductors*, (Springer-Verlag, Berlin, 1984).
- <sup>26</sup>M. J. R. Hoch and R. J. Hoch, Phys. Rev. B **51**, 16016 (1995).
- <sup>27</sup>L. P. Gor'kov and P. L. Krotkov, Phys. Rev. B **67**, 033203 (2003).
- <sup>28</sup>K. V. Kavokin, Phys. Rev. B **64**, 075305 (2001).