

## Effect of Paramagnetic Impurities on Nuclear Spin-Lattice Relaxation: A Numerical Study

C. Ebner

*Department of Physics, The Ohio State University, Columbus, Ohio 43210\**  
*Battelle Memorial Institute, Columbus, Ohio 43201*

C. C. Sung<sup>†</sup>

*Department of Physics, The University of Alabama in Huntsville, Huntsville, Alabama 35807*  
 (Received 28 June 1973)

The effect of paramagnetic impurities on the nuclear spin-lattice relaxation time in metals is investigated by applying the theory of Blume and Hubbard in an appropriately generalized form. The calculations are carried out at finite temperature and magnetic field strength assuming that the Ruderman-Kittel-Kasuya-Yosida interaction couples the impurity spins. The effect of approximating this interaction by  $1/r^3$  is examined as is the temperature, magnetic field, and impurity-concentration dependence of the relaxation time. Quantitative results are obtained using parameters appropriate to the rare-earth alloy  $\text{La}_{1-c}\text{Gd}_c\text{Al}_2$  and a comparison is made with experiment; agreement is generally good.

### I. INTRODUCTION

Time-dependent spin correlations are of great importance in magnetic resonance and relaxation phenomena. Blume and Hubbard<sup>1</sup> recently developed a general formalism for calculating the time dependence of the correlation function of a system of spins coupled via the Heisenberg exchange interaction. We have used<sup>2</sup> this formalism to calculate the magnetic field dependence of the spin-lattice relaxation time  $T_1$  in solid  $\text{He}^3$ ; measurements<sup>3</sup> show that  $T_1$  is proportional to  $e^{-\omega/\omega_0}$  at large  $\omega$ , where  $\omega$  is the Larmor frequency of the spins in the applied field and  $\omega_0$  is a constant. Our calculations also give this field dependence which is not found in any of the phenomenological approaches to relaxation phenomena.<sup>4</sup>

The theory of Blume and Hubbard may also be used to explain how paramagnetic impurities affect nuclear-spin relaxation. It has long been known from experiments<sup>5</sup> that a relative concentration  $c$  of paramagnetic impurities changes the inverse spin-lattice relaxation time by an amount  $\Delta(1/T_1) \sim c^{-1/2}$  for  $c$  close to 1 and by an amount  $\Delta(1/T_1) \sim 1/c$  for  $c \ll 1$ ; in the latter regime, the spin-correlation function is a Lorentzian. These experimental facts, as well as some other novel features of the time dependence of the correlation function, may be derived by applying the theory of Ref. 1 with suitable modifications to take the randomness of the impurity positions into account.<sup>6</sup> The results are consistent with the experiments of McHenry *et al.*<sup>7</sup> on  $\text{La}_{1-c}\text{Gd}_c\text{Al}_2$  alloys and with some earlier work on a similar system.<sup>8</sup>

The model in Ref. 6 is typical of those used in studies of the effect of paramagnetic impurities on the nuclear spin-lattice relaxation time; the relaxation of the nuclear spins is enhanced through their dipole-dipole (d-d) interaction with the para-

magnetic ions which are considered to be randomly distributed and which couple via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. There is much speculation<sup>9</sup> concerning the nature of the interaction between the nuclear spin and that of the paramagnetic impurity; the treatment given in Ref. 6 is sufficiently general that it may be extended to other types of interaction rather trivially.

There are several approximations in Ref. 6 which have been improved upon in the present work in order to make a quantitative comparison with experiment. First, when calculating the nuclear spin-lattice relaxation time in Ref. 6, the angular part of the d-d interaction was replaced by a constant; this was believed to alter the magnitude of the result but not its functional dependence on any physical parameters. Second, the oscillations in the RKKY interaction were discarded in Ref. 6 so that the paramagnetic ions interact via a  $1/r^3$  law where  $r$  is the interimpurity distance. This approximation is commonly used<sup>10</sup> but is certainly of questionable validity when the concentration of impurities is not too small. Finally, at finite (as opposed to infinite) temperature we should improve the calculations of Ref. 6 by distinguishing between the longitudinal and transverse parts of the spin-correlation function.<sup>11</sup> The extension to finite  $T$  introduces some important corrections to results which were previously given in the literature<sup>7</sup> and which are based on the phenomenological theory. In connection with this point, see also Ref. 9.

The remainder of this paper is organized as follows: In Sec. II, we review briefly the formalism of Blume and Hubbard and also derive equations for the longitudinal and transverse parts of the spin-correlation function at finite  $T$ . Section III contains a discussion of the results and a comparison with the experiments<sup>7</sup> on  $\text{La}_{1-c}\text{Gd}_c\text{Al}_2$ .

Finally, in Sec. IV, we demonstrate the power of the formalism by indicating how it may be applied to the physically very different problem of NMR studies on orthohydrogen (o-H<sub>2</sub>) impurities in solid parahydrogen (p-H<sub>2</sub>). In this system, the intra d-d interaction of the o-H<sub>2</sub> molecules is influenced by a much stronger electric quadrupole-quadrupole interaction<sup>12</sup> which varies as  $1/r^5$ .

## II. THEORETICAL BACKGROUND

The quantity that we wish to calculate is the spin-correlation function of nuclear spins  $\tilde{I}_i$  located at positions  $\tilde{r}_i$  which interact with paramagnetic impurity spins  $\tilde{S}_j$  at  $\tilde{r}_j$  via a dipole-dipole interaction<sup>4,13</sup>

$$H_{d-d} = \sum_{i,j} \frac{\gamma_i \gamma_j}{r_{ij}^3} \left( \tilde{I}_i \cdot \tilde{S}_j - \frac{3(\tilde{I}_i \cdot \tilde{r}_{ij})(\tilde{S}_j \cdot \tilde{r}_{ij})}{r_{ij}^2} \right), \quad (1)$$

where  $\tilde{r}_{ij} = \tilde{r}_i - \tilde{r}_j$  and  $\gamma_i$  ( $\gamma_s$ ) is the gyromagnetic ratio of the nuclear (impurity) spins.<sup>14</sup> The Zeeman energy of the spins in an external magnetic field  $\tilde{H}_0 = H_0 \hat{z}$  is

$$H_z = -\gamma_I H_0 \sum_i I_i^z - \gamma_S H_0 \sum_j S_j^z, \quad (2)$$

where the superscript  $z$  (or  $x$  or  $y$ ) on the spin operator denotes the Cartesian component.

In computing the spin-correlation function of the impurities, we may neglect  $H_{d-d}$  because it is much weaker than the interaction between two paramagnetic spins; this is taken to be the RKKY interaction,

$$H_{\text{RKKY}} = -\frac{J'}{2} \sum_{i,j} \frac{\tilde{S}_i \cdot \tilde{S}_j}{(2k_F r_{ij})^3} \times \left( \cos 2k_F r_{ij} - \frac{\sin 2k_F r_{ij}}{2k_F r_{ij}} \right). \quad (3)$$

Here  $k_F$  is the Fermi momentum of the electron liquid in the host material and  $J'$  is the coupling constant in the RKKY interaction,

$$J' = 8\pi\epsilon_F [N(\epsilon_F)J]^2,$$

where  $\epsilon_F$  is the Fermi energy  $N(\epsilon_F)$  the density of states at the Fermi surface, and  $J$  the strength of the conduction-electron-impurity spin-exchange coupling.

In what follows, we use a rotating frame of reference so that  $S^* e^{i\omega_s t}$  becomes simply  $S^*$  in our equations; also, we subtract  $\langle S^* \rangle$  from  $S_i^z$  and relabel the difference  $S_i^z$ . Here  $\langle S^* \rangle$  is the ensemble average of the  $z$  component of the impurity spin operator and  $\omega_s = \gamma_s H_0$  is the Larmor frequency of the impurity spins;  $S^*$  are the usual spin raising and lowering operators.

Our procedure is, first, to compute the normalized longitudinal (transverse) dynamical response function  $G_i^{(z)}(t)$  of the paramagnetic im-

purities,

$$G_i^{(z)}(t) \equiv \langle \delta S_i^{(z)}(t) \rangle / \langle \delta S_i^{(z)}(0) \rangle, \quad (4)$$

where  $\delta S_i$  is the response to a unit external field,

$$\delta S_i^{(z)}(t) = i \int_{-\infty}^0 [S_i^{(z)}(t), S_i^{(z)}(t')]_{-} e^{\epsilon t'} dt'; \quad (5)$$

$[\ , ]_{-}$  denotes the commutator. It may be shown<sup>6</sup> that  $\delta S_i^{(z)}(t)$  is proportional to the Fourier transform of  $\chi''(\tilde{r}_i, \omega)/\omega$ ,  $\chi''$  being the dynamical susceptibility.<sup>15</sup>

In the short-time approximation,<sup>16</sup>  $\delta \tilde{S}_i(t)$  satisfies the operator equation<sup>1,17</sup>

$$\delta \dot{S}_i = i \tilde{h}_i(t) \times \tilde{S}_i(t),$$

where

$$\tilde{h}_i(t) = -\sum_j J(r_{ij}) \tilde{S}_j(t) \quad (6)$$

and

$$J(r) = \frac{J'}{(2k_F r)^3} \left( \cos 2k_F r - \frac{\sin 2k_F r}{2k_F r} \right).$$

Further,  $G_i^{(z)}$  may be written as

$$G_i^z(t) = \exp \left[ - \left( \sum_j J^2(r_{ij}) \int_0^t dt_1 \int_0^{t_1} dt_2 \times \langle S_j^z(t_1) S_j^z(t_2) + S_j^y(t_1) S_j^y(t_2) \rangle \right) \right] \quad (7)$$

and

$$G_i^x(t) = \exp \left[ - \left( \sum_j J^2(r_{ij}) \int_0^t dt_1 \int_0^{t_1} dt_2 \times \langle S_j^x(t_1) S_j^x(t_2) + S_j^y(t_1) S_j^y(t_2) \rangle \right) \right]. \quad (8)$$

These equations must be solved simultaneously. First, we introduce the fluctuation-dissipation theorem to convert the correlation functions into  $G^z$  and  $G^x$ ; following Hubbard,<sup>18</sup> we write

$$\langle S_j^z(t) S_j^z(0) \rangle = T \chi^z G_j^z(t) \quad (9)$$

and

$$\langle S_j^x(t) S_j^x(0) \rangle = T \chi^x G_j^x(t), \quad (10)$$

where  $T \chi^z = S^2 \partial B_s(x) / \partial x$  and  $T \chi^x = S^2 B_s(x) / x$ ; here  $B_s(x)$  is the Brillouin function of order  $S$  and  $x = \gamma_s S H_0 / T$ . Equations (9) and (10) are valid when  $T$  is higher than the magnetic transition temperature of the spins; Eqs. (7) and (8) reduce to the corresponding equation of Ref. 1 when the high-temperature approximation is used,  $B_s(x)/x = \frac{1}{3} S(S+1)$ .

We next apply a cumulant expansion to Eqs. (7) and (8) in order to average over the random variable  $r_{ij}$ ; assuming the system is homogeneous so that the average of  $G_i$ , called  $\bar{G}$ , is independent of  $i$ , we obtain

$$\bar{G}^z(t) = \exp \left( -c \int_{r>\alpha} \frac{d^3 r}{a^3} (1 - e^{-2J^2(r)y^z(t)}) \right) \quad (11)$$

and

$$\bar{G}^t(t) = \exp\left(-c \int_{r>a} \frac{d^3r}{a^3} (1 - e^{-J^2(r)} [y^t(t) + y^t(t')])\right). \quad (12)$$

Here  $a^3$  is the volume per molecule in the crystal; also

$$y^t(t) = S^2 \frac{B_s(x)}{x} \int_0^t dt' (t-t') \bar{G}^t(t') \quad (13)$$

and

$$y^t(t) = S^2 \frac{\partial B_s(x)}{\partial x} \int_0^t dt' (t-t') \bar{G}^t(t'). \quad (14)$$

Some higher-order terms in  $c^2$  have been neglected in Eqs. (11) and (12); consequently, they are only applicable at small impurity concentrations. However, when  $c$  becomes of order 1, the important part of  $\bar{G}^t(t)$  is at small  $t$  such that  $J(a)t \leq 1$ , in which regime  $\bar{G}^t(t) \sim e^{-cJ^2(a)t^2}$ . In this range of  $t$  the (neglected) higher-order terms appear as a series in powers of  $cJ^2(a)t^2$ . They are not qualitatively different from the leading term so far as the  $c$  dependence is concerned; thus Eqs. (11) and (12) may be more useful than anticipated at large impurity concentrations.

The quantity which is actually measured is the time dependence of the fluctuations in the nuclear spin or  $g^{I(t)}(t) \equiv \langle \delta I^{I(t)}(t) \rangle / \langle \delta I^{I(t)}(0) \rangle$ , where we assume homogeneity and drop the subscript identifying a particular spin. These functions can be found using exactly the same approach as before. The d-d interaction between two nuclear spins is much smaller than that between a nuclear spin and an impurity; so we may keep only the latter and find, for  $\bar{g}^t(t)$ ,

$$\bar{g}^t(t) = \exp\left(-c \int_{r>a} \frac{d^3r}{a^3} (1 - e^{-(a/r)^6 y_2(\theta, \varphi, t)})\right), \quad (15)$$

where

$$y_2 = \omega_D^2 S^2 \int_0^t dt' (t-t') \left( \bar{G}^t(t') \vartheta |F_1(\theta, \varphi)|^2 \frac{\partial B_s(x)}{\partial x} + \bar{G}^t(t') \cos(\omega_s t')^{1/2} \times [9 |F_2(\theta, \varphi)|^2 + |F_0(\theta, \varphi)|^2] \frac{B_s(x)}{x} \right). \quad (16)$$

Further,  $\omega_D = \gamma_I \gamma_s / a^3$ ,  $F_0 = 1 - 3 \cos^2 \theta$ ,  $F_1 = \sin \theta \cos \theta e^{i\varphi}$ , and  $F_2 = \sin^2 \theta e^{2i\varphi}$ .

### III. RESULTS AND APPLICATIONS

Equations (11)–(14) have been solved numerically to obtain  $\bar{G}^t$  and  $\bar{G}^t$  as functions of  $zt$  where  $z = SJ^2 / (2k_F a)^3$ . The impurity concentration  $c$  lies in the interval  $0.1 \geq c \geq 0.001$  while  $x$  ranges from 0.5 to 7 and  $k_F a = 6.43$ . The range of  $c$  is the same as in the experimental work of Ref. 7 while  $k_F a$  has the value appropriate to  $\text{LaAl}_2$  assuming three conduction electron per atom and a spherical Fermi

surface. The procedure used in solving is simply to iterate  $zt$ , starting from  $\bar{G}^{t(t)}(0) = 1$ ; this is quite easy since the right-hand sides of Eqs. (11) and (12) depend only on  $\bar{G}^{t(t)}(zt')$  at arguments  $t' \leq t$ . The iterations are carried to  $zt$  sufficiently large that both correlation functions are smaller than  $10^{-4}$ .

From  $\bar{G}^{t(t)}$ , we find  $\bar{g}^t$  by performing the integration indicated in Eqs. (15) and (16); for each  $x$  and  $c$  this has been done at values of  $H_0$  such that  $3.6 \leq H_0 \leq 24.4$  kOe. The  $H_0$  were chosen to coincide with the fields used by McHenry *et al.*<sup>7</sup>

Our results generally confirm those given in Ref. 6; some new features are produced by distinguishing between  $\bar{G}^t$  and  $\bar{G}^t$  and by improving the numerical approximations. The major points are as follows.

The time dependence of  $\bar{G}^{t(t)}$  may be summarized as Gaussian at small  $t$ , simple exponential at intermediate  $t$ , and finally  $\sim e^{-(t/\tau)^{1/2}}$  at large  $t$ ; the regime in which we find these behaviors are unchanged from Ref. 6 and will not be described further here.

For small  $x$  (high- $T$  limit),  $\bar{G}^t \equiv \bar{G}^t$  as expected from the fact that  $B_s(x)/x = \partial B_s(x) / \partial x$  in this limit. As  $x$  becomes larger,  $\bar{G}^t$  is smaller than  $\bar{G}^t$ . A typical case is shown in Fig. 1 where  $\bar{G}^t$  and  $\bar{G}^t$  are plotted against  $zt$  for  $c = 0.01$  and  $x = 7$ ; the nature of the time dependence is explicitly shown in Fig. 2 where the logarithms of the same functions are given as functions of  $zt$  (curves A) and as func-

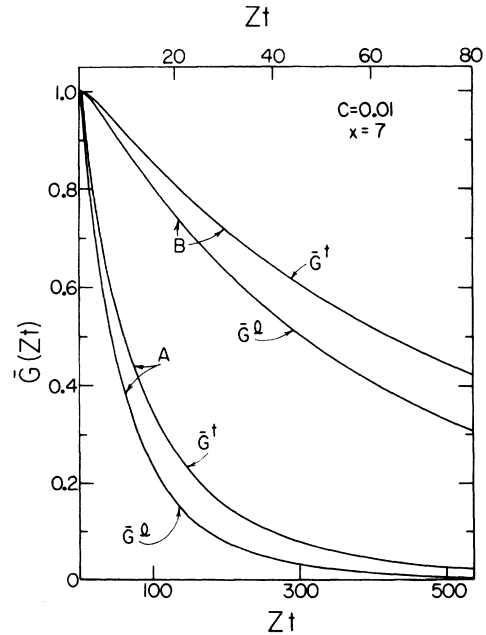


FIG. 1.  $\bar{G}^t$  and  $\bar{G}^t$  vs  $zt$  for  $c = 0.01$ ,  $x = 7$ . Curves A (B) are referred to the lower (upper) abscissa.

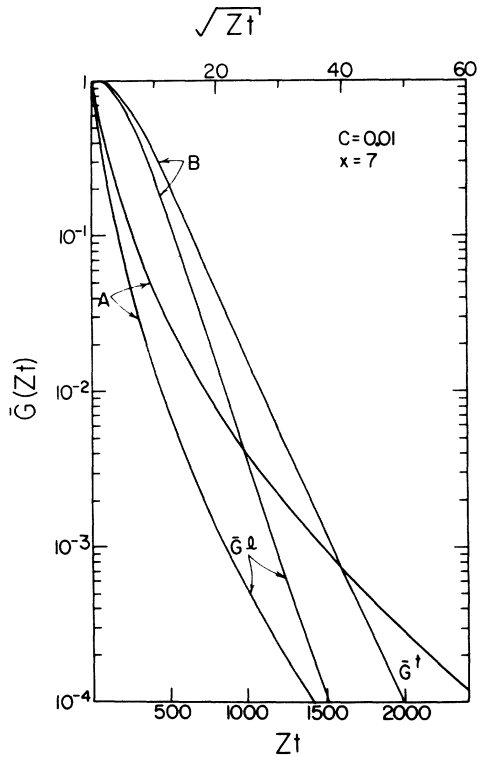


FIG. 2.  $\log_{10}(\bar{G}^A)$  and  $\log_{10}(\bar{G}^B)$  plotted vs  $zt$  (curves A refer to lower abscissa) and vs  $(zt)^{1/2}$  (curves B refer to upper abscissa) for  $c=0.01$ ,  $x=7$ .

tions of  $(zt)^{1/2}$  (curves B). Note that the Gaussian behavior exists over such a short interval of  $zt$  that it cannot be observed in this figure. The simple exponential behavior, seen as a straight line in curves A, dominates until  $\bar{G} \approx 0.3$ ; for larger  $t$ ,  $\bar{G} \sim e^{-(t/\Gamma)^{1/2}}$  which appears as a straight line in curves B.

In the limit of large  $t$ , we find  $\bar{g}^A(t) \sim e^{-(t/\Gamma_0)^{1/2}}$  in agreement with Ref. 6 where the angle dependence of  $y_2$  was treated in an approximate way. The concentration dependence of  $1/\Gamma_0$  is demonstrated in Fig. 3 for several different values of  $H_0$  and  $x$ . At small  $H_0$ , there is strong deviation from purely linear behavior at all  $x$  for  $c$  not too small. This effect may be traced to the integral

$$\int_0^t dt' (t-t') \bar{G}^A(t') \cos \omega_s t'$$

in Eq. (16); at large  $H_0$ ,  $\omega_s$  is large and the cosine factor makes the integral average to 0. If  $H_0$  is small, the cosine is essentially 1, giving a large integral. The  $c$  dependence arises from the fact that  $\bar{G}^A(t')$  decays more slowly as  $c$  is decreased; thus decreasing  $c$  has the same effect as increasing  $H_0$ . Notice that in the conventional analysis of experimental data,  $\omega_s \Gamma \gg 1$  is always assumed.

For given  $x$  and  $c$ , the value of  $1/\Gamma_0$  may be

varied to some extent by varying  $H_0$ ; if  $H_0$  is constrained to lie within certain limits (e.g.,  $3.6 \leq H_0 \leq 24.4$  kOe), then  $1/\Gamma_0$  may be varied by some amount  $\Delta(1/\Gamma_0)$ . This spread in  $1/\Gamma_0$  depends on  $x$  and  $c$  and also on the parameter

$$z = \frac{8\pi\epsilon_F [N(\epsilon_F)J]^2 S}{(2k_F a)^3}$$

Unfortunately,  $z$  is not known with any precision; as discussed by McHenry *et al.*,<sup>7</sup> experimentally inferred values of  $J$  alone vary between 0.057 and 0.11 eV (we used  $J=0.087$  eV) so that  $z$  may reasonably be altered by a factor of 2. For the fields used in Fig. 3, this change of  $z$  produces a change in  $\Delta(1/\Gamma_0)$  by about the same factor. The importance of this point is brought out by Figs. 4(a) and 4(b) where  $1/\Gamma_0$  is shown as a function of  $x$  and the results compared with the experiments reported in Ref. 7. At the larger concentrations ( $c=0.1, 0.05$ ) there is reasonable agreement between the calculated and measured spreads in  $1/\Gamma_0$ , but as  $c$  decreases, the calculated  $\Delta(1/\Gamma_0)$  is much smaller than what is measured. The agreement can be improved somewhat by using a different value of  $z$ ; on the other hand, at least some of the experimental spread in  $\Delta(1/\Gamma_0)$  seems to be produced by uncertainty in the data. Consequent-

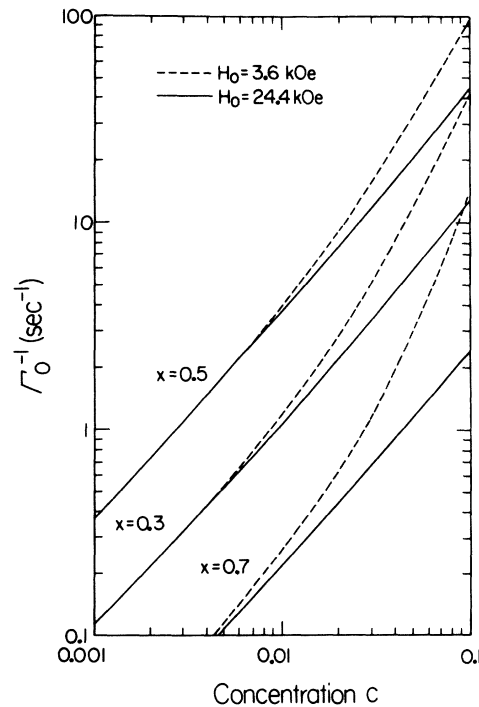


FIG. 3. Theoretical inverse nuclear-spin relaxation time  $1/\Gamma_0$  ( $\text{sec}^{-1}$ ) as a function of impurity concentration  $c$  for values of  $x=\gamma_s S H_0/T$  equal to 0.5, 3, and 7 and for fields  $H_0=3.6$  and 24.4 kOe.

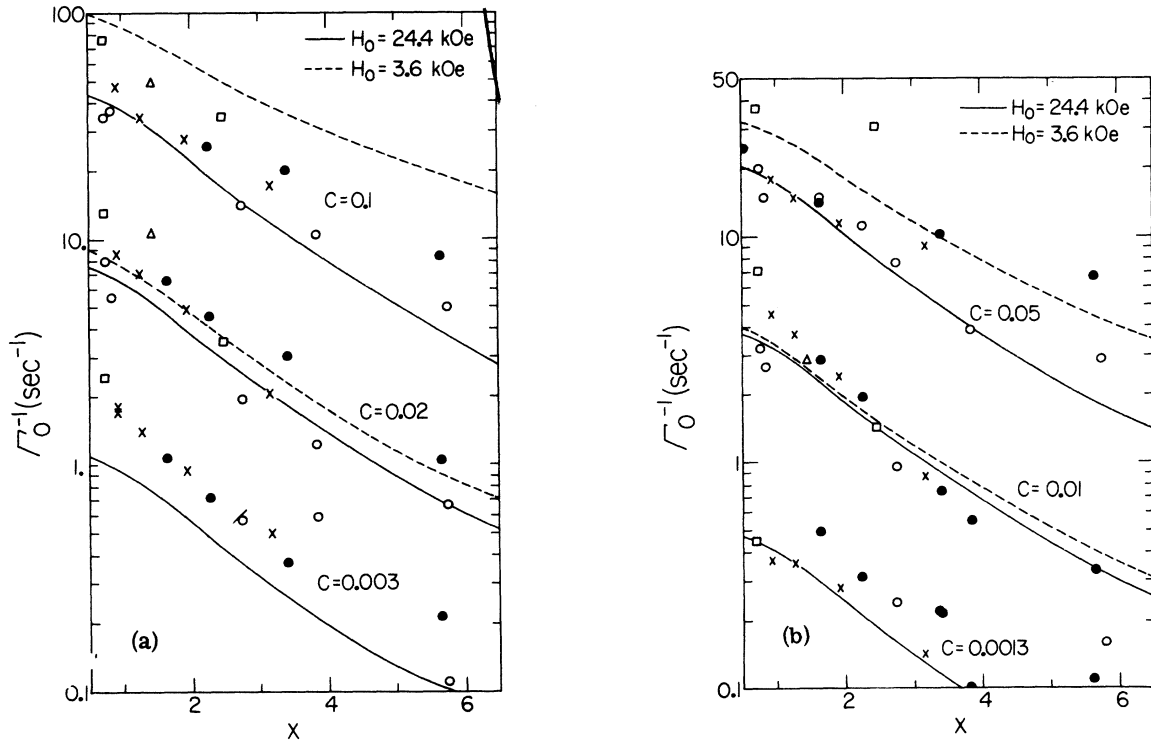


FIG. 4. Inverse nuclear-spin relaxation time  $1/\Gamma_0$  ( $\text{sec}^{-1}$ ) vs  $x = \gamma_s S H_0 / T$ ; the theoretical curves are for  $c = 0.1, 0.05, 0.02, 0.01, 0.003$ , and  $0.0013$  at magnetic fields  $H_0 = 3.6$  kOe. The experimental data are taken on samples with the same concentration of Gd at fields of 3.6 ( $\Delta$ ), 6.3 ( $\square$ ), 8.1 ( $\times$ ), 14.4 ( $\bullet$ ), and 24.4 kOe ( $\circ$ ).

ly, we have not tried to improve the fit of  $\Delta(1/\Gamma_0)$  by adjusting  $z$ .

We have, however, adjusted the magnitude of  $1/\Gamma_0$  by picking  $\omega_D$  to give the best agreement of theory and experiment. If  $\omega_D$  is evaluated using parameters appropriate to  $\text{La}_{1-c}\text{Gd}_c\text{Al}_2$ , it turns out to be about a factor of 2 larger than the value we employed. This does not necessarily indicate an inadequacy in our treatment of the nuclear spin-impurity interaction; we could have obtained the same magnitude of  $1/\Gamma_0$  by using  $\omega_D$  appropriate to  $\text{La}_{1-c}\text{Gd}_c\text{Al}_2$  and by taking  $z$  a factor of 2 larger, corresponding to  $J = 0.12$  eV. We may conclude, then, that it is possible to obtain quite good agreement of theory and experiment without using any unreasonable values of the various parameters that enter the theoretical expressions.

Finally, we have also examined the effect of approximating the RKKY interaction by simpler expressions. In particular, the factor  $\cos 2k_F r - (\sin 2k_F r)/2k_F r$  has been replaced by (a)  $\cos 2k_F r$  and by (b) 1. Case (a) produces almost no change in any of the results. The reason is that in Eqs. (11) and (12), the behavior of  $J^2(r)$  at large  $r$  ( $2k_F r \gg 1$ ) is responsible for cutting off the integration over  $d^3r$ , so the  $(\sin 2k_F r)/2k_F r$  correction is not important. For the same reason, our use

of the continuum approximation in place of the sum over lattice sites has no effect on the results.

Approximation (b), on the other hand, in which  $\cos 2k_F r$  is replaced by 1, does introduce some quantitative changes. The function  $J^2(r)$  is in effect made stronger by this substitution so that the correlation functions  $\bar{G}^{(t)}(t)$  decay more rapidly, leading to a decrease in  $1/\Gamma_0$  by some 35–40%. Qualitatively, however, there is no change in the various time, concentration, field, and temperature dependence of the various quantities. This suggests that the oscillations in the RKKY interaction serve only to reduce its effective strength and that the actual period of the oscillations is unimportant so long as it is short compared to distances over which  $r$  varies in Eqs. (11) and (12) before the integrand goes to zero. We have tested this idea by changing the period of the oscillations by an appreciable ( $\sim 20\%$ ) amount. As expected, the effect is quite negligible.

#### IV. SOLID MIXTURES OF ORTHOHYDROGEN AND PARAHYDROGEN

We have also computed the spin-lattice relaxation time of o- $\text{H}_2$  and p- $\text{H}_2$  mixtures as a function of o- $\text{H}_2$  concentration  $c$ . To this end, we assume the o- $\text{H}_2$  molecules interact via a potential  $J(r)$

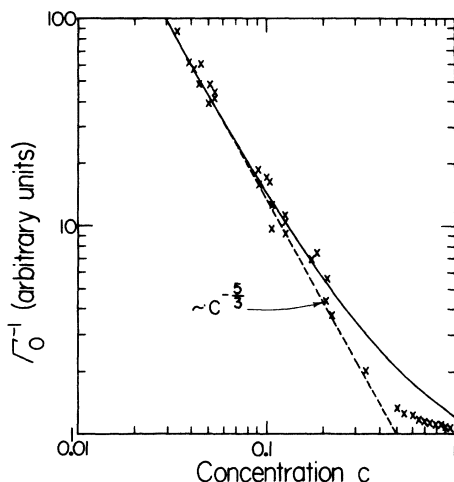


FIG. 5. Solid curve gives the impurity dependence of the inverse spin-lattice relaxation time  $1/\Gamma_0 = 2\int_0^\infty \bar{G}(t) dt$  in arbitrary units. The dashed line with slope  $-\frac{5}{3}$  is asymptotic to the theoretical curve for  $1/\Gamma_0$  at low impurity concentrations. Experimental data are also shown for solid mixtures of o-H<sub>2</sub> in p-H<sub>2</sub> as a function of o-H<sub>2</sub> concentration  $c$ .

$= J(a/r)^5$  where  $J$  and  $a$  are constants and  $r$  is the intermolecular distance. The relaxation time is

given by  $1/\Gamma_0 \sim I$ , with  $I = 2\int_0^\infty \bar{G}(t) dt$ ,  $\bar{G}$  being the impurity-averaged spin-correlation function of the o-H<sub>2</sub>. In Fig. 5, the logarithm of  $1/\Gamma_0$  is plotted against the logarithm of  $c$ . We find that the slope of this line approaches the value of  $-\frac{5}{3}$  for  $c$  very small and decreases monotonically as  $c$  increases, in general agreement with the data which are taken from Fig. 4 of Ref. 12. The magnitude of the theoretical inverse lifetime has been adjusted to give the best agreement of theory and experiment at intermediate concentrations  $c \sim 0.1$ .

This calculation has been included here only to demonstrate the general applicability of our approach; it is quite incomplete in that replacement of the electric quadrupole-quadrupole interaction by  $1/r^5$  is a poor approximation. Also, when  $c$  is very small, the crystal field plays an important role. A detailed discussion of this problem will be given elsewhere.

#### ACKNOWLEDGMENTS

The authors wish to thank Dr. B. G. Silbernagel for more detailed plots of the data reported in Ref. 7. They also acknowledge the use of the Ohio State University Computing Center's IBM 370/65 facility.

\*Permanent address.

†Work supported in part by a NASA Grant under Contract No. NGL-01-008-001.

<sup>1</sup>M. Blume and J. Hubbard, Phys. Rev. B **1**, 3815 (1970).

<sup>2</sup>C. C. Sung and C. Ebner, Solid State Commun. **11**, 1659 (1972).

<sup>3</sup>See, e.g., W. C. Thomlinson, J. F. Kelly, and R. C. Richardson, Phys. Lett. A **38**, 531 (1972), and references contained therein.

<sup>4</sup>See, e.g., A. Abragam, *The Principles of Nuclear Magnetism* (Oxford U.P., London, 1961).

<sup>5</sup>For a review of earlier work, see J. Hama, T. Inuzuka, and T. Nakamura, Prog. Theor. Phys. **48**, 1768 (1972).

<sup>6</sup>C. C. Sung and L. G. Arnold, Phys. Rev. B **7**, 2095 (1973).

<sup>7</sup>M. R. McHenry, B. G. Silbernagel, and J. H. Wernick, Phys. Rev. B **5**, 2958 (1972).

<sup>8</sup>D. Tse and S. R. Hartman, Phys. Rev. Lett. **25**, 510 (1968).

<sup>9</sup>For a review, see A. Narath, Crit. Rev. Solid State Sci. **3**, 1 (1972).

<sup>10</sup>For example, in calculating the heat capacity of the random

impurities. See M. W. Klein, Phys. Rev. **173**, 552 (1968).

<sup>11</sup>There are several different approaches in the literature. See W. Götze and P. Wölfle, J. Low Temp. Phys. **5**, 575 (1971); and R. E. Walstedt and A. Narath, Phys. Rev. B **6**, 4118 (1972); C. C. Sung, Phys. Rev. B (to be published).

<sup>12</sup>For a review of the experimental NMR studies in solid hydrogen, see F. Weinhaus and H. Meyer, Phys. Rev. B **7**, 2974 (1973).

<sup>13</sup>We use units such that  $\hbar = k_B = 1$ .

<sup>14</sup>All physical parameters used will be those appropriate to the La<sub>1-c</sub>Gd<sub>c</sub>Al<sub>2</sub> system; a useful table of such parameters is included in Ref. 7.

<sup>15</sup>P. C. Martin and L. P. Kadanoff, Ann. Phys. (N.Y.) **24**, 419 (1963).

<sup>16</sup>The meaning and justification of this approximation are explored in Refs. 1 and 6. We emphasize that this approximation should be checked for each system.

<sup>17</sup>F. B. McLean and M. Blume, Phys. Rev. B **7**, 1149 (1973).

<sup>18</sup>J. Hubbard, J. Phys. C **4**, 53 (1971).