case of indirect excitons[9] this interaction is not forbidden by translational symmetry. It may be realized in the form of simultaneous scattering of electrons and holes between different X points by either Coulomb or electron-phonon interaction.

A quantitative determination of the matrix elements in question and the deformation potentials is difficult because of uncertainties in the decomposition of overlapping bands A, B, and C. At the present time we can only give a rough estimate of these quantities. Using the hydrostatic deformation potential[10] to eliminate Δ_1 we find that $\Delta_3 \approx 7$ eV per unit shear deformation $\frac{2}{3}e_{zz} - \frac{1}{3}(e_{xx}+e_{yy})$. This rather large value again indicates that the exciton transition does not take place at the center of the Brillouin zone but that a star degeneracy is involved. Preliminary analysis shows that $E_1 < E_2 < E_3$ with $E_3 - E_1 \approx 3$ meV and that the scattering matrix element M_{iv} is about 1 meV. The same scattering mechanism presumably also holds for TlBr where a similar but smaller splitting of the first exciton band is observed.[2]

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Impurity-Induced Nuclear Spin-Lattice Relaxation in Metals: A New Approach

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We find an $\exp[-(t/\tau_1)^{1/2}]$ time dependence for the recovery of the Al^{27} nuclear magnetization in the system $La_{1-c}Gd_cAl_2$. This is characteristic of diffusionless relaxation and an especially direct interpretation of the data is possible in this limit. Variation of τ_1 with temperature and magnetic field indicates that longitudinal dipolar fluctuations of the Gd moments relax the Al^{27} nuclei while its concentration dependence indicates Ruderman-Kittel-Kasuya-Yosida coupling between the Gd spins.

We have studied the Al^{27} nuclear spin-lattice relaxation in the system $La_{1-c}Gd_cAl_2$. Conditions are such in these materials that contributions to relaxation from the Gd impurities can be interpreted in a way which does not depend on the poorly defined quantities[1] usually encountered in impurity relaxation studies: the diffusion constant, the diffusion barrier radius, and the observation barrier radius. This considerable simplification permits an unambiguous determination of the impurity relaxation process operative in metals which we will compare with other recent work.[2-4]

A number of interactions between the nuclei

and impurities can result in spin-lattice relaxation.[5] The angular averaged nuclear relaxation rate, $1/T_1(r)$, can be calculated as a function of distance r from an impurity by use of standard perturbation techniques. If nuclear spin diffusion is absent and the impurities are dilute, $c \ll 1$ where c is the ratio of magnetic to nonmagnetic rare earth sites, the magnetization $M_z(t)$ will approach equilibrium as[6]

$$p(t) = \exp\{-4\pi N_0 c \int_{r_0}^{\infty} [1 - e^{-t/T_1(r)}] r^2 dr\}, \quad (1)$$

where $p(t) = 1 - M_z(t)/M_z(\infty)$, $M_z(\infty)$ is the equilibrium magnetization, and $N_0 c$ is the number of magnetic sites per unit volume. Nuclei lying closer to an impurity than the observation barrier radius r_0 will be shifted out of the observed resonance line and are therefore excluded from consideration. This expression leads to an exponential time dependence only for short times: $t \ll T_1(r_0)$. This limit is used in interpreting many of the previous studies of impurity relaxation in metals.[2-4] The relaxation rate in this limit is given by

$$1/T_1 = 4\pi N_0 c \int_{r_0}^{\infty} r^2 dr / T_1(r). \quad (2)$$

If relaxation results from dipolar or Ruderman-Kittel-Kasuya-Yosida (RKKY)[7] interactions, [which behave as $1/T_1(r) = \mathcal{C}/r^6$], one finds

$$1/T_1 = \tfrac{4}{3}\pi N_0 c \mathcal{C}/r_0^3. \quad (3)$$

Our main concern is the limit $t \gg T_1(r_0)$, for in this case Eq. (1) has the form[6]

$$p(t) \propto \exp\{-(t/\tau_1)^{1/2}\}, \quad (4)$$

where

$$(1/\tau_1)^{1/2} = \tfrac{4}{3}\pi^{3/2} N_0 c \mathcal{C}^{1/2}. \quad (5)$$

The relaxation rate *does not depend on* r_0 and consequently we can study the details of the relaxation process itself, as embodied in $\mathcal{C}$. This is fortuitous, since r_0 is poorly defined, and the value of $1/T_1$ depends very sensitively on the choice of its value. Further, r_0 has a strong temperature and field dependence at low temperatures where impurity polarization is appreciable. Since a relaxation process is identified by studying the field and temperature dependence of $1/T_1$ at low temperatures, the variation in r_0 will complicate the analysis of such variations in $\mathcal{C}$.[8]

Several conditions must be met for this asymptotic behavior to be observable. First, diffusion must be negligible, a condition which occurs in $LaAl_2$ presumably because of quadrupole decoupling of the Al spin-spin interaction. The very long spin-spin relaxation time ($T_2 = 600$ μsec, as compared to ~20 μsec in Al metal) inhibits diffusion.[9] Second, magnetization recovery from sources other than the impurities must be slow enough to observe the limit $t \gg T_1(r_0)$. Since Korringa[10] relaxation with conduction electrons is the dominant process in $LaAl_2$, this means that the Korringa rate $1/T_{1K}$ must be sufficiently small so that $T_{1K} > T_1(r_0)$. The Korringa process is an order of magnitude weaker for Al^{27} in $LaAl_2$ [$1/T_{1K}T = 0.07$ (K sec)$^{-1}$] than the Cu, Cd, and Sc hosts used in previous studies. The simultaneous presence of slow diffusion and weak spin-lattice relaxation in $LaAl_2$ provides a unique opportunity to investigate this long-time behavior.

The recovery of the Al^{27} nuclear magnetization has been studied using spin-echo techniques as a function of c (= 0.0013, 0.003, 0.01, and 0.02), magnetic field H (= 3.6, 6.3, 8.1, 14.4, and 24.4 kOe), and temperature T (= 1.2, 2.0, 3.0, 4.2, 14.0, and 20.4 K). The transmitter was modified[11] to saturate the quadrupolar broadened line (width ~1.4 MHz) of the powdered samples by sweeping the rf frequency during a train of several hundred pulses.

The magnetization of pure $LaAl_2$ recovers at $\exp(-t/T_{1K})$, while we observe a recovery of the Gd-doped samples described by

$$1 - \frac{M(t)}{M(\infty)} \propto \exp\left[-\left(\frac{t}{T_{1K}}\right) - \left(\frac{t}{\tau_1}\right)^{1/2}\right]. \quad (6)$$

This behavior is shown for a typical set of data in Fig. 1, where we have multiplied each datum point by $\exp(+t/T_{1K})$ to remove the conduction-electron contribution to the relaxation.[12] This $t^{1/2}$ decay is observed for all samples throughout the temperature and field range discussed here.

An analysis of these τ_1 data enables us to determine the type of impurity-nuclear coupling. Figure 2 shows a plot of $1/\tau_1$ for the $c = 0.003$ and $c = 0.02$ samples as a function of the parameter $1/x = k_B T/gS\mu_B H$, where μ_B is the Bohr magneton, $g = 2$ for Gd, $S = \frac{7}{2}$ is the spin, and k_B is the Boltzmann constant. The curves drawn through the data represent the function

$$\frac{1}{\tau_1} = 1.4 \times 10^3 c \frac{\partial B_{7/2}(x)}{\partial x} \text{ sec}^{-1}, \quad (7)$$

where $B_{7/2}(x)$ is the Brillouin function of spin $\frac{7}{2}$. This dependence of $1/\tau_1$ on $\partial B_{7/2}(x)/\partial x$ is characteristic of spin-lattice relaxation caused by longitudinal fluctuations of the impurity's dipolar

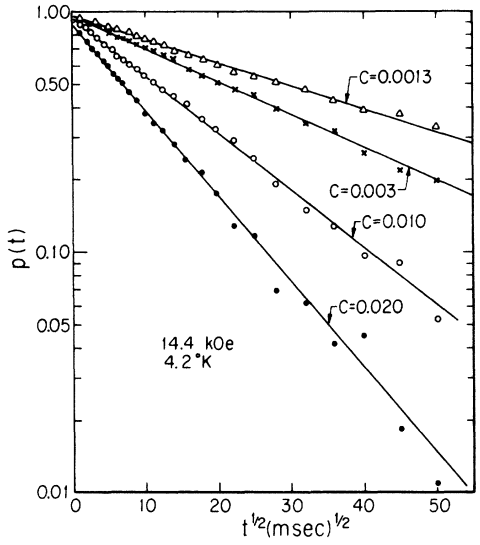

FIG. 1. A plot of the impurity contribution $p(t)$ to the magnetization recovery, versus $t^{1/2}$. The experimental points $1-M(t)/M(\infty)$ are multiplied by $\exp(t/T_{1K})$ to eliminate the effects of Korringa relaxation. The linear dependence of $\ln p(t)$ vs $t^{1/2}$ indicates the existence of a diffusionless relaxation process.

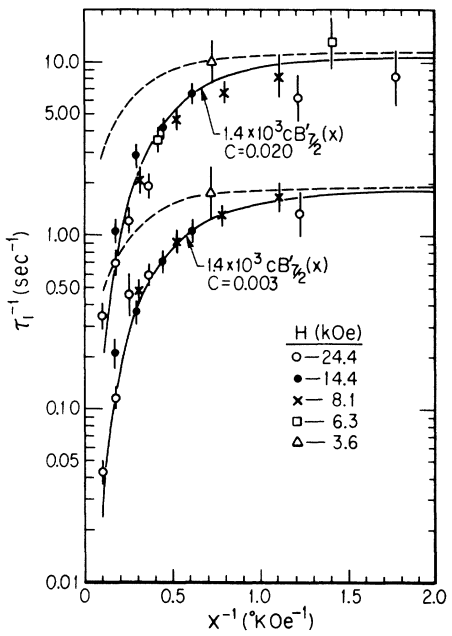

FIG. 2. A plot of the impurity relaxation rate $(1/\tau_1)$ vs x^{-1}, where $x=g\mu_B SH/k_B T$, for temperatures between 1.2 and 20°K and the magnetic fields indicated. Included are the functions $\partial B_{7/2}(x)/\partial x$ (solid line) and $B_{7/2}(x)/x$ (dashed line) expected for various impurity relaxation mechanisms.

field[5,13] and easily distinguished from the $B_{7/2}(x)/x$ dependence, plotted for comparison, that is appropriate for relaxation from transverse fluctuations.[5] If these fluctuations decay exponentially with a characteristic time τ_m, the form of $1/T_1(r)$ becomes

$$\frac{1}{T_1(r)} \equiv \frac{\mathcal{C}}{r^6} = \frac{1}{r^6}\frac{6}{5}(\gamma_m\gamma_n\hbar)^2 S^2 \frac{\partial B_s(x)}{\partial x}\frac{\tau_m}{1+(\omega_n\tau_m)^2}, \quad (8)$$

where γ_m and γ_n are the gyromagnetic factors of the Gd impurities and the nuclei, respectively, and ω_n is the nuclear Larmor frequency. From Eqs. (5) and (8), with $N_0 = 1.48\times10^{22}$ cm^{-3}, we find the theoretical value [assuming $(\omega_n\tau_m)^2 \ll 1$]

$$\left(\frac{1}{\tau_1}\right)_{\text{theor}} = 2.96\times10^{15}\tau_m c^2 \frac{\partial B_{7/2}(x)}{\partial x}\ \text{sec}^{-1}. \quad (9)$$

Comparing this expression with the experimental result [Eq. (7)] we can solve for

$$1/\tau_m = 2.1\times10^{12} c\ \text{sec}^{-1}. \quad (10)$$

The decay of the impurity correlations could arise from either spin-lattice relaxation to the conduction electrons or an interaction between the impurities. However, the impurity spin-lattice relaxation rate $[\pi J^2 N^2(0) k_B T/\hbar = 4\times10^8 T$ sec^{-1}] is too slow to account for the observed value of $1/\tau_m$, and the linear dependence of $1/\tau_m$ suggests that interactions between the impurities are responsible for this decay. The Gd spins are primarily coupled by an RKKY interaction,[7] which in the free-electron approximation has the form

$$H_{ij}^{\text{RKKY}} = 4\pi E_F N^2(0) J^2 F(2k_F r_{ij})\vec{S}_i\cdot\vec{S}_j, \quad (11)$$

where $N(0)$ is the density of states at the Fermi surface per atom for both spin directions (= 0.47 states/eV atom), E_F is the Fermi energy (= 9.5 eV), k_F is the Fermi momentum, J is the average conduction-electron–Gd exchange energy ($H_{ex} = -2J\vec{S}\cdot\vec{s}$), and $F(x) = (x\cos x - \sin x)/x^4$ is the RKKY range function. A short-time expansion of the correlation function has been made in the high-temperature limit with the following approximations: (1) The correlation function is assumed to be a Gaussian; (2) we average the range function $F^2(2k_F r) \simeq \frac{1}{2}(2k_F r)^{-6}$; and (3) a configuration average over the random impurity sites is made.[6] With these approximations we find an average correlation function that is exponential and has a decay rate given by

$$\frac{1}{\tau_m} \simeq \frac{1}{9}\left(\frac{\pi S(S+1)}{6}\right)^{1/2}\frac{N(0)J^2 c}{\hbar}. \quad (12)$$

In the free-electron model one finds agreement with the result of $1/\tau_m$ deduced from experiment [Eq. (10)] if $|J|=0.1$ eV. This value of J is in reasonable agreement with results of a free-electron model interpretation of the superconducting transition temperature suppression ($|J|=0.073$ eV) found by Maple[14] in magnetically dilute $La_{1-c}Gd_cAl_2$.

We believe that this is the first observation of the asymptotic $t^{1/2}$ limit of diffusionless relaxation in a metal. The simplifying factors we have discussed enable us to identify unambiguously the impurity-process relaxation as arising from longitudinal dipolar fluctuations. Although the functional dependence is complicated in previous studies[2-4] by the possible temperature variation of τ_0, we find that the magnitude and functional dependence of the high-field ($\omega_m\tau_m>1$) data in each of these cases can be accounted for by the same mechanism.[15]

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Self-Consistent Theory of Clusters in Disordered Alloys

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A way to deal with cluster effects in disordered alloys is given through an extension of the coherent-potential-approximation theory. An explicit expression is derived for pair effects and is shown to give the exact c^2 term in the lower concentration limit.

The coherent-potential approximation (CPA) is known to be what might be called the best single-site approximation in the disordered-alloy problem. Our purpose here is to generate higher approximations which give self-consistent solutions for pairs, triplets, etc. Some attempts along these lines have recently been made through various techniques[1-4]; as we shall notice later on, they have failed to give the correct behavior in the low-concentration limit. This point will be fully discussed elsewhere.

Here we give an extension of the formalism developed by Velický, Kirkpatrick, and Ehrenreich[5] in the single-site approximation to take into account pairs of atoms or larger clusters. We start from the usual Hamiltonian H describing an alloy $A_{1-c}B_c$:

$$H = H_0 + V, \tag{1}$$

with

$$H_0 = \sum_{n\neq m} |n\rangle\beta_{nm}\langle m|, \tag{2}$$

$$V = \textstyle\sum_n |n\rangle\epsilon_n\langle n|, \tag{3}$$