

FIG. 1. Spin-orbit splitting diagram of  ${}^4T_{2g}(D)$  level in  $Mn^{2+}$  for  $\xi$  between 0 to 400  $cm^{-1}$ ,  $B=840$ ,  $C=3080$ ,  $D_4=780$ , and  $\alpha=76$   $cm^{-1}$ . The observed fine-structure components of the level are marked  $\Phi$  at  $\xi=320$   $cm^{-1}$ .

respectively. The calculated spin-orbit splitting of  ${}^4T_{2g}(D)$  level obtained by diagonalizing the matrices on an IBM-7044 computer is drawn in Fig. 1 for  $\xi$  values ranging from 0 to 400  $cm^{-1}$ . First-order degeneracy of  $\Gamma_6$  and  $\Gamma_8'$  levels is found to be removed, but the levels still lie quite close to one another. Therefore, three spin-orbit components are expected to be observed.

Figure 2 shows the observed structure at 77°K. Four prominent peaks marked  $D_1$ ,  $D_2$ ,  $D_3$ , and  $D_{1v}$  are seen, whose energies in  $cm^{-1}$  are 27 917, 28 129, 28 258, and 28 385. Analysis of the separations between the lines suggests that the lines  $D_1$ ,  $D_2$ , and  $D_3$  are probably the three expected fine-structure components. The separations  $D_3-D_2$  and  $D_2-D_1$  are in the ratio of 129:212  $\approx$  3:5, which is in agreement with the first-order splitting ratio. For find-

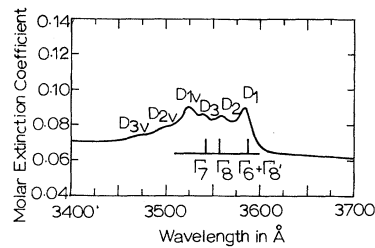


FIG. 2.  ${}^6A_{1g}(S) \rightarrow {}^4T_{2g}(D)$  band in  $RbMnF_3$  at 77°K. The calculated spin-orbit components of  ${}^4T(D)$  level for  $\xi=320$   $cm^{-1}$  are marked for comparison.

ing the  $\xi$  value we considered the total splitting  $D_3-D_1=341 \pm 10$   $cm^{-1}$ , and the corresponding  $\xi$  value is  $320 \pm 10$   $cm^{-1}$ . Comparison between the calculated and observed components is given in Figs. 1 and 2, and the agreement is seen to be fairly satisfactory.

The line  $D_{1v}$  probably belongs to another vibrational level of  ${}^4T_{2g}(D)$  state. From the similarity in the shapes of  $D_1$  and  $D_{1v}$  it appears as if  $D_{1v}$  is the first spin-orbit component in that vibrational state. In that case there should be two more lines on the higher energy side of  $D_{1v}$ . Indications of the presence of two such lines can be seen in Fig. 2, where these are marked as  $D_{2v}$  and  $D_{3v}$ . However, because of the very low intensities of these lines no measurements could be performed.

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## SPIN-LATTICE RELAXATION OF NUCLEAR SPIN ECHOES IN METALS

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Measurements of nuclear magnetic relaxation times have been reported for a number of ferromagnetic metals and alloys, and, where data for both longitudinal ( $T_1^{-1}$ ) and transverse spin-echo ( $T_2^{-1}$ ) relaxation rates are given, it has consistently been found that  $T_2'^{-1} > T_1^{-1}$ , where  $T_2'^{-1}$  is the contribution to  $T_2^{-1}$  from spin-lattice interactions. For example, ratios

$T_1/T_2'$  equaling 3, 5, and approximately 2 have been reported for the systems  $Ni^{61}$  in  $Ni$ ,<sup>1</sup>  $Mn^{55}$  in  $Fe$ ,<sup>2</sup> and  $Mn^{55}$  in  $Ni$ ,<sup>3</sup> respectively, where  $T_1^{-1}$ ,  $T_2'^{-1} \propto T$ . Explanations for these puzzling results have been offered<sup>2,4</sup> in terms of anisotropies in the hyperfine field fluctuations that cause the relaxation. This approach, however, is unsatisfactory for several reasons,

among them the existence of a predominant and essentially isotropic contribution to  $T_1^{-1}$  from orbital fluctuations.<sup>5,6</sup>

Here, we propose instead that this phenomenon is a consequence of strong first-order quadrupolar broadening of the nuclear Zeeman levels for<sup>7</sup>  $|m| > \frac{1}{2}$ . The spin-echo signal therefore arises mainly from the  $m = \pm\frac{1}{2}$  levels and decays at a (spin-lattice) rate of the order of  $W_{-1/2,1/2}$ , which is greater than  $T_1^{-1}$  for nuclear values  $I > \frac{1}{2}$ . Assuming magnetic dipole relaxation, we have  $W_{m, m+1} = W_0[I(I+1) - m(m+1)]$  with<sup>8</sup>

$$T_1^{-1} = 2W_0 \quad (1a)$$

and

$$T_2'^{-1} \sim W_{-1/2,1/2} = W_0(I + \frac{1}{2})^2. \quad (1b)$$

We expect  $T_1/T_2'$ , then, to be a rapidly increasing function of  $I$ , in clear accord with the relaxation data for various systems summarized in Table I. We note that the  $V_{0.01}Rh_{0.99}$  and  $Co_{0.02}Rh_{0.98}$  systems with the largest ratio  $T_1/T_2' \sim 12$  are nonmagnetic. Relaxation data for these materials are reported here for the first time.

The condition that the entire spin-echo signal comes from the  $m = \pm\frac{1}{2}$  energy levels leads to the largest ratio  $T_1/T_2'$  observable for a given value of  $I$ . It is of interest to calculate this maximum ratio and compare it with experiment. This we may do with a slightly modified density-matrix approach. Consider an ensemble of isolated nuclear spins in a metal-

lic system subjected to a field  $H_0$ . The density matrix  $\rho$  for such a system in thermal equilibrium will be diagonal in the  $I_z$  representation with matrix elements  $\langle m | \rho_0 | m \rangle = a(1 + m\epsilon)$ , where  $\epsilon = \gamma\hbar H_0/kT \ll 1$  and  $a^{-1} = \text{Tr}(1)$ . It is most convenient to work this problem in a reference frame  $(x', y', z')$  rotating at a frequency  $\omega_0 = \gamma H_0$  about the  $z$  axis. To study the transverse decay we apply a pulse of field  $H_1$  along the  $y'$  axis (at the exact resonance frequency). The assumption of quadrupolar broadening is incorporated now by allowing only the matrix elements  $\langle \pm\frac{1}{2} | I_y | \mp\frac{1}{2} \rangle$  to cause transitions in  $\rho'$ . The effects of the rf pulse are thus limited to the  $m = \pm\frac{1}{2}$  manifold of  $\rho'$ , giving it the form

$$\rho'(t_w) |_{m=\pm\frac{1}{2}} = \begin{bmatrix} 1 & \frac{1}{2}\epsilon \\ \frac{1}{2}\epsilon & 1 \end{bmatrix}$$

after a pulse of duration  $t_w = \pi/[\gamma H_1(2I+1)]$ . The remaining portion of  $\rho'$  is unchanged. Such a pulse produces a transverse magnetization  $M_{x'} = \frac{1}{4}N_0\gamma\hbar\epsilon$ , which then decays irreversibly at a rate  $T_2'^{-1}$  to be determined (dephasing effects from static inhomogeneous broadening do not affect the echo decay and are ignored).

To calculate the decay of  $M_{x'}$ , we rediagonalize  $\rho'(t_w)$  by introducing new basis functions and then write down a set of rate equations in terms of populations. This is accomplished with the set of functions  $\varphi_{\pm} = (1/\sqrt{2})(\varphi_{1/2} \pm \varphi_{-1/2})$  and  $\varphi_m$  for  $|m| > \frac{1}{2}$ . In this representation

$$\rho'(t_w) |_{\pm} = \begin{bmatrix} 1 + \frac{1}{2}\epsilon & 0 \\ 0 & 1 - \frac{1}{2}\epsilon \end{bmatrix}$$

Table I. Spin-lattice relaxation data for various metals and alloys.

Material	Nucleus	$I$	$T_1T$ (sec°K)	$T_2'T$ (sec°K)	$(T_1/T_2')$ expt	$(T_1/T_2')$ max
$V_{0.01}Rh_{0.99}$	$V^{51}$	$\frac{7}{2}$	2.6	0.22	11.8	12.25
$Co_{0.02}Rh_{0.98}$	$Co^{59}$	$\frac{7}{2}$	$3.9 \times 10^{-2}$	$3.0 \times 10^{-3}$	13.0	12.25
$Mn_{0.01}Fe_{0.99}$	$Mn^{55}$	$\frac{5}{2}$	$4.5 \times 10^{-2}$	$9 \times 10^{-3}$	$\sim 5$	7.00
$Mn_{0.01}Ni_{0.99}^a$	$Mn^{55}$	$\frac{5}{2}$	$2.86 \times 10^{-2}$	$1.37 \times 10^{-2}$	2.1	7.00
$Ni^a$	$Ni^{61}$	$\frac{3}{2}$	$5.55 \times 10^{-2}$	$1.82 \times 10^{-2}$	3.3	3.25
Pt	$Pt^{195}$	$\frac{1}{2}$	$2.98 \times 10^{-2}$	$2.89 \times 10^{-3}$	0.97	1.00
$Fe^b$	$Fe^{57}$	$\frac{1}{2}$	...	...	$\sim 1$	1.00

<sup>a</sup>The relaxation times for these systems have been found to be field dependent [Ref. 3, and V. Jaccarino, K. Kaplan, R. E. Walstedt, and J. H. Wernick, Phys. Letters 23, 514 (1966)], attaining a constant value for  $H_0 \gg \frac{4}{3}\pi M$ . The high-field values are quoted here.

<sup>b</sup>Weger's data (Ref. 1) for  $Fe^{57}$  was taken for nuclei in the domain walls. Thus, although  $T_1T$  was not constant,  $T_1$  and  $T_2$  were found to be indistinguishable.

as expected. We define a set of populations  $n_{\pm}$  and  $n_m$ ,  $|m| > \frac{1}{2}$ , in the new representation and note that  $M_{x'}(\text{observed}) \propto n_{\pm} - n_{-}$ . The decay of  $M_{x'}$  is then governed by

$$\frac{d}{dt}(n_{+} - n_{-}) = -(W_{+, -} + W_{-, +})(n_{+} - n_{-}) - n_{+}(W_{+, 3/2} + W_{+, -3/2}) \\ + n_{-}(W_{-, 3/2} + W_{-, -3/2}) + n_{3/2}(W_{+, 3/2} - W_{-, 3/2}) + n_{-3/2}(W_{+, -3/2} - W_{-, -3/2}). \quad (2)$$

The Boltzmann factors for the various  $W$ 's lead only to second- and higher-order effects in  $\epsilon$  and are therefore omitted.

The various  $W$ 's in Eq. (2) may be calculated by assuming a spin-lattice interaction of the form  $\mathcal{H}_{sl} = -\gamma(H_x I_x + H_y I_y + H_z I_z)$ . The quantities  $H_{\eta}$  represent the fluctuating part of hyperfine field contributions from all sources and are in general quantum-mechanical operators. We assume that the  $H_{\eta}$  have correlation time  $\tau_c \ll \omega_0^{-1}$  and that they have identical spectral densities (i.e., isotropic fluctuations) when averaged over the electronic states of the system. The transition rate between a pair of nuclear states ( $i, j$ ) is then simply given by

$$W_{ij} = 2W_0[|\langle i | I_x | j \rangle|^2 + |\langle i | I_y | j \rangle|^2 + |\langle i | I_z | j \rangle|^2], \quad (3)$$

where all information concerning the properties of the "lattice" is embodied in  $W_0 \propto T$ . This definition coincides with that of  $W_{m, m+1}$  given above.

The rates in (2) may immediately be evaluated with (3) to give

$$W_{+, -} = W_{-, +} = \frac{1}{2}W_0[I(I+1) + \frac{7}{4}]$$

and

$$W_{+, 3/2} = W_{+, -3/2} = W_{-, 3/2} \\ = W_{-, -3/2} = \frac{1}{4}W_0[I(I+1) - \frac{3}{4}].$$

After substitution, Eq. (2) assumes the form

$$\frac{d}{dt}(n_{+} - n_{-}) = -(1/T_2') \max(n_{+} - n_{-})$$

with

$$(1/T_2') \max = W_0 \left[ \frac{3}{2}I(I+1) + \frac{7}{8} \right], \quad (4)$$

which is reasonably close to the estimate in Eq. (1b). The theoretical ratio

$$(T_1/T_2') \max = \frac{3}{4}I(I+1) + \frac{7}{16}$$

is tabulated with the data in Table I and is seen to be in good agreement in almost all cases. It is somewhat remarkable, for example, that the  $\text{Co}^{59}$  and  $\text{V}^{51}$  results for  $T_1/T_2'$  are in such

close accord, since the  $\text{Co}^{59}$  relaxation rates are strongly enhanced, presumably by local Coulomb effects, and are two orders of magnitude faster than those of the nearly identical nucleus of  $\text{V}^{51}$ . In the case of  $I = \frac{1}{2}$  there is only one rate in the problem and we must have  $T_2' = T_1$ , as was found by Butterworth<sup>8</sup> for  $\text{Pt}^{195}$  in Pt and by Weger<sup>1</sup> for  $\text{Fe}^{57}$  in Fe.

To the extent that the quadrupolar broadening is not large (compared with  $H_1$ ), there will be increased contributions to the echo signals from higher  $m$  levels and correspondingly longer times  $T_2'$  approaching the limit  $T_2' = T_1$  for the case of no broadening at all. On this basis one might guess that for the  $\text{Mn}^{55}$  in Ni studies,<sup>3</sup> the participation of  $|m| = \frac{3}{2}, \frac{5}{2}$  levels in the echo signals was nearly complete. This phenomenon leads in principle to an  $H_1$  dependence of  $T_2'$ . Preliminary results show roughly a 40% increase in  $T_2'$  for  $\text{V}^{51}$  in  $\text{V}_{0.005}\text{Rh}_{0.995}$  on increasing  $H_1$  from 6.5 to 13.8 G, as one might expect from including a wider portion of the spectrum in the echo. This sort of study can provide a simple estimate of quadrupolar broadening in disordered alloys and other systems.

A simple confirmation of the assumption of extreme quadrupolar broadening is provided by the enhanced " $\gamma$ " one finds when applying a  $\frac{1}{2}\pi$  pulse only to the  $\pm\frac{1}{2}$  levels of a spin system. The above calculation shows that the requisite pulse "area"  $H_1 t_w$  is reduced by a factor  $(I + \frac{1}{2})$  in this case. Thus, it was found for the  $\text{V}_{0.01}\text{Rh}_{0.99}$  system ( $I + \frac{1}{2} = 4$  for  $\text{V}^{51}$ ) that the  $\frac{1}{2}\pi$  pulse time was comparable with that of protons in glycerine under the same conditions, the  $\gamma$  for protons being roughly 4 times that of  $\text{V}^{51}$ .

Finally, it must be commented that, although the mechanism proposed here is very likely to be operative in the systems shown in Table I, it does not rule out the possibility of "real" anisotropies in spin-lattice interaction for these materials. However, such effects can only be isolated by first carefully accounting for the present phenomenon.

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<sup>7</sup>We limit ourselves to the case of odd half-integral spin here.

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## INVESTIGATION OF LANDAU-DAMPING EFFECTS ON SHOCK FORMATION

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Landau damping in plasmas of equal ion and electron temperatures (alkali plasmas) may prevent the formation of a shock. Shocks are produced when the ratio  $T_e/T_i$  is increased to about 8 or so by cooling the ions through  $i-n$  collisions.

We report in this note preliminary results concerning the formation of steep wave fronts (shocks) in a  $Q$  device.<sup>1</sup> A schematic view of the experimental arrangement is shown in Fig. 1(a). The plasma is produced by surface ionization of cesium atoms on a hot ( $\sim 2500^\circ\text{K}$ ) tantalum plate, and is confined radially by a uniform and constant magnetic field of intensity up to 10 000 G. The plasma column, about 1 m long, is terminated at the opposite end from the generating plate by a second tantalum plate, which can either be heated up to  $\sim 2500^\circ\text{K}$  or left at room temperature. A grid is inserted at 30 cm from the generating plate, the plane of the grid being normal to the  $\vec{B}$  lines. The grid consists of tungsten wires  $2.5 \times 10^{-3}$  cm in diameter, spaced  $3 \times 10^{-2}$  cm. It is normally biased at  $-20$  V with respect to the generating plate, and it absorbs most of the ions from the plate, thereby producing a plasma density distribution along the axis as indicated in Fig. 1(b). By suddenly varying the grid bias to approximately  $-2$  V, the grid is "open," i.e., its transmission to the plasma is strongly enhanced. Our measurements indicate that the voltage pulse on the grid may vary its transmission from  $\sim 10$  to  $\sim 80\%$ . A similar technique has already been used<sup>2</sup> to study the propagation and damping of ion-acoustic waves in the cesium plasma of a  $Q$  device. In the work of Ref. 2, however, the density modulation by the grid amounted only to a few percent of its dc value and was sinusoidal in time. In our present ar-

angement the "opening" of the grid is somewhat the equivalent of the breaking of the diaphragm in a conventional shock tube. It should be noticed, however that (a) our experiments are performed in alkali plasmas produced in a manner entirely independent of the presence of a shock, and (b) the ion and the electron temperatures,  $T_i$  and  $T_e$ , are approximately equal ( $\sim 2500^\circ\text{K}$ ) if the pressure of the neutral gas in the device is kept sufficiently low ( $p \lesssim 10^{-4}$  mm Hg). At higher neutral gas pressures one may cool the plasma ions to about room temperature while keeping  $T_e$  near  $2500^\circ\text{K}$ , thereby achieving values of  $T_e/T_i$  as large as 8 or so.

In a first series of experiments we have investigated, by means of Langmuir probes movable along the plasma column, the propagation

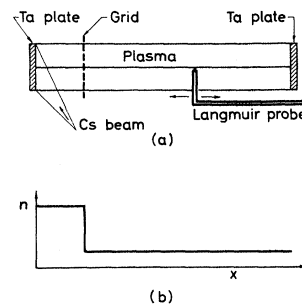


FIG. 1. (a) Schematic diagram of the experimental arrangement, and (b) density distribution along the plasma column before "opening" of the grid.