where

$$x=2(v/c)(E_\gamma/\Gamma),\quad p=2\mu_e H/I_e\Gamma,\quad q=2\mu_g H/I_g\Gamma,$$

and

$$N(x;p,q)=N(-x;pq)=N(x;-p,-q),\ N(0;0,0)=1.$$

For $Ni^{61}(I_e=\frac{5}{2}, I_g=\frac{3}{2})$ one gets 35 different pairs of (m_e-m_e') and (m_g-m_g') values, that is, a combination of 35 Mössbauer lines. The unknown parameters are H and μ_e or p and q. We have written a code for the ORACLE computer, which yields $N(x)$ for fixed p and q values in the form of a curve. Many of these curves have the measured half-width $x_{\frac{1}{2}}=2E_\gamma(v_{\frac{1}{2}})_{\text{exp}}/c\Gamma=8.75$ and a shape in agreement with our measurements (one typical case is shown in Fig. 4).

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Electron Spin-Lattice Relaxation of Chromium in MgO†

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The effective spin-lattice relaxation time, T_1, has been observed for the ground-state multiplet of Cr(+3) in the cubic field of MgO. T_1 is observed to vary from 800 milliseconds at 1.3° to 3.4 milliseconds at 50°K. The direct process dominates below 4.2°.

SPIN relaxation within the ground-state multiplet of the chromium (+3) ion has been of interest for some time.[1–8] We have observed the spin-lattice relaxation of Cr(+3) in the cubic environment of MgO from 1.3°K to 50°K. For this case, i.e., a multiplet having equally spaced energy levels and therefore a common spin temperature, a unique spin-lattice time is expected and was found in very dilute samples. The general problem of spin relaxation in MgO at even moderate concentrations is complicated apparently by the existence of other impurities.

The paramagnetic resonance spectra of Cr(+3) substituted for Mg(+2) in MgO have been clearly shown[9] to be due to some chromium ions in a cubic environment and to others having an Mg(+2) vacancy nearby. On the basis of charge neutrality, the concentration of excess Mg(+2) vacancies is expected to be at least half that of trivalent impurities. Apparently, a considerable fraction of the trivalent impurities are closely associated with Mg(+2) vacancies. The central line from a Cr(+3)−[100] vacancy pair lies about 2 gauss above the cubic chromium line at 9 kMc/sec with H along [100].

The microwave spectra of the other transition elements in MgO have been reported.[9,10,11] The crystals being used in the present study show absorption by Cr(+3), Fe(+3), Mn(+2), V(+2), and Fe(+2) in a cubic environment, and Cr(+3)-vacancy pairs. At 9 kMc/sec, one of the Mn(+2) $(-\frac{1}{2}-\frac{1}{2})$ lines is about one gauss below the cubic chromium line, the $(-\frac{1}{2}-\frac{1}{2})$ line of Fe(+3) with H along [100] is about 50 gauss below, and the central pair of V(+2) lines are split to about 40 gauss each side of the cubic chromium line. The Fe(+2) line at 1800 gauss[10] was observed only in some of the samples which had higher chromium concentration. Spectrographic analysis showed impurity concentrations (other than for chromium) to be of the order of 10 parts per million (ppm), with Fe consistently higher than the others.

Using the inversion-recovery method previously described,[4] the spin population difference, n, is observed directly as a function of time after inversion by adiabatic fast passage. When recovery of equilibrium for $S=\frac{1}{2}$ is dominated by spin-lattice relaxation, the deviation of n from its equilibrium value is expected to vary as $\exp(-t/T_1)$, where T_1 is the spin-lattice relaxation time. This single exponential recovery is also expected for Cr(+3) in the cubic environment of MgO. Conversely, the observation of nonexponential recovery of the cubic chromium line implies that other relaxation processes are operative.

Exponential recovery of the cubic Cr(+3) line was observed in two of the purest crystals available as a function of temperature up to 50°K; the values of T_1 so obtained are listed in Table I. For spin-lattice relaxation by resonant phonons[1] at frequency f, T_1

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TABLE I. Spin-lattice relaxation time for Cr(+3) in MgO with $H \approx 3$ Kg. Values of T_1 are given in milliseconds to an accuracy of 10%.

Temperature	50°K	4.2°K	2.1°K	1.4°K	1.3°K
Sample A[a]	3.8	300	550	740	···
Sample B[b]	3.4	···	560	···	800

[a] Grown at the General Electric Company; kindly supplied by N. S. Shiren.

[b] Grown at SemiElements Incorporated, Saxonburg, Pennsylvania.

should vary with temperature as $[1+\exp(-x)]/[1-\exp(-x)]$, where $x=hf/kT$. For small x, T_1 is then expected to be proportional to $1/T$. The temperature dependence of T_1 is observed to be $1/T$ from 1.3°K to 4.2°K for which temperature range x is less than one for all the transitions within the Cr(+3) multiplet. Therefore, the direct process dominates spin-lattice relaxation in this range. Measurements of the temperature dependence at a larger value of f/T would be necessary to determine which of the transitions are effective. The more rapid decrease of T_1 between 4.2°K and 50°K suggests that spin relaxation is dominated at 50°K by multiple phonon process.

The order-of-magnitude formulas by Mattuck and Strandberg[3] give the following:

1. T_1 for the direct process to be about two orders of magnitude longer than the observed value of 300 milliseconds at 4.2°K.
2. T_1 for the Raman process to be close to the observed value of 3 milliseconds at 50°K.

A check on the relaxation model used in reference 3 requires a more precise calculation of transition probabilities from their general expressions; such a comparison is underway.

It was found that the Cr(+3) resonance, with a full width at half-power absorption of about 0.5 gauss, was very inhomogeneous in these samples. A "hole" inverted[4] in the line persisted for a time of the order of T_1; the source of the broadening is not yet understood but is under further study. Inspection of the wings of the line gave no unambiguous evidence of hyperfine coupling to Mg^{25} as had been observed in the F-center spectra.[12] Dipolar coupling to paramagnetic impurities other than cubic Cr(+3) and anisotropic hyperfine coupling to Mg^{25} are sources of some inhomogeneity.

The impurity concentration in sample B was determined by spectrographic analysis to have within a factor of three the following values (in ppm): Cr-15, Fe-80, Mn-2, V-5; Ti-5; Ni-10, Co<10. The concentrations of Cr(+3) and Mn(+2) were found by resonance absorption to be nearly equal in A and also in B. The linewidths of the Fe(+3), Mn(+2), and V(+2) lines with H along [100] are closely the same as that for the Cr(+3) line. Fe(+2) was not observed in these samples, even though the apparent Fe(+3) concentrations are far below that indicated for Fe by spectrographic analysis.

It is interesting to note that the recovery from inversion observed for the cubic Cr(+3) line in many other crystals is nonexponential and more rapid. The asymptotic time constant[4] is observed to be as short as 300 microseconds at 2°K in crystals containing up to 500 ppm chromium and about the same concentration of other transition elements as is present in A and B. The line is broader and even homogeneous in the crystals showing the faster recoveries. Further study is in progress, but significant results are likely to require much better crystals.

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