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Spin-lattice Relaxation in Cerium Magnesium Nitrate at Helium Temperatures: A new process *

by C. B. P. Finn, R. Orbach ** and W. P. Wolf

We have made measurements of the spin-lattice relaxation time, T_1, of a single crystal of $Ce_2Mg_3(NO_3)_{12}.24H_2O$, using the method of Casimir and du Pré [1], at frequencies between 175 c/s and 900 c/s and d.c. fields of 300, 600 and 1000 oersteds. In all fields an extremely rapid variation of T_1 with temperature was observed, (see Fig. 1), T_1 increasing by a factor of about 350 between 3°K and 1.9°K. Outside this temperature range the limited frequencies available to us at the time prevented quantitative measurements, but the trend indicated that T_1 varied much less rapidly between 1.9°K and 1.1°K. Above 3°K, T_1 seemed to continue its rapid variation, because no paramagnetic resonance absorption was observed above 10°K even at 10^{10} c/s [2].

To interpret these results we have concentrated on measurements at relatively high fields; for these it is a good approximation to neglect spin-spin interaction effects compared with the Zeeman splittings [2]. There are no complications from nuclear hyperfine structure or small Stark splittings $\sim$ kT, and, under these conditions, the salt behaves very much like an assembly of ideal $S = \frac{1}{2}$ spins save for an anisotropic g-value. There is, however, one important additional factor: specific heat [3] and susceptibility [4] measurements have shown that there is an excited energy state ($|\, J_z = \pm 3/2\rangle$) relatively close to the ground doublet ($\Delta/k = 34°K$) and we suggest that this dominates the relaxation between the components of the ground state doublet in the temperature range covered by our experiments.

In the usual treatment of spin-lattice relaxation [6], as applied to salts of the iron group, one considers excited states at energies which are much larger than the maximum phonon energy $\sim k\theta_D$. Hence there are no

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simple spin-phonon processes with energy sufficient to cause real transitions to the excited states, and relaxation proceeds by second and higher order processes involving spin-orbit and orbit-lattice interactions. However, if $\Delta \ll k\theta_D$ [6], real phonon-induced transitions to the excited states become possible and relaxation of the ground state doublet can occur by a two stage process, each *step* of which is a single phonon (or ' direct ') process [7]. (See Fig. 2). At temperatures at which $kT \ll \Delta$, T_1 due to such a mechanism should vary as $e^{\Delta/kT}$. Comparison with the experimental results (Fig. 1) shows that this is exactly what is observed between 3°K and 2°K. The slopes of the lines of log T_1 versus 1/T are 34 ± 1°K, in good agreement with the value $\Delta/k \simeq 34$°K deduced from specific heat and susceptibility.

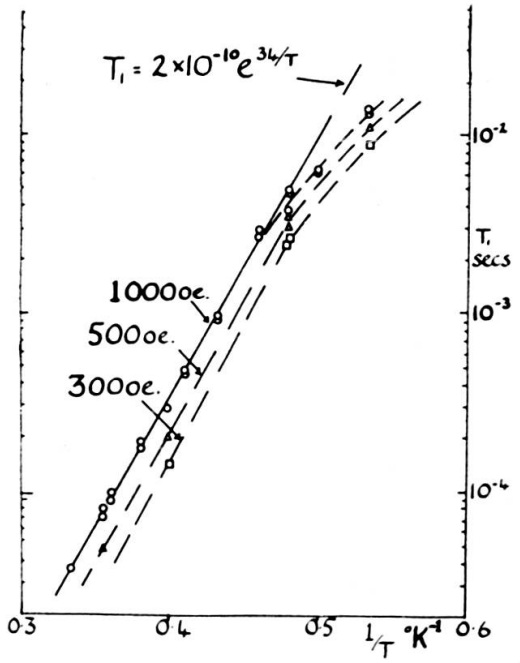

Fig. 1.

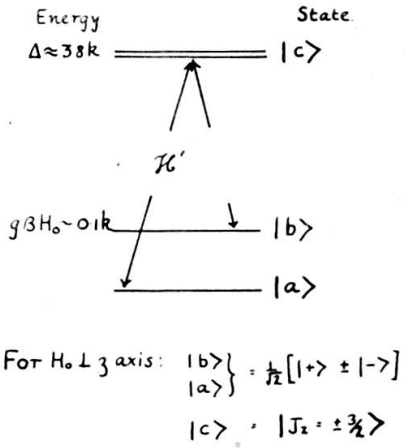

Fig. 2.

The absolute magnitudes of relaxation times in rare earth salts are in general very short, since the magnetic states are functions of $J = L + S$, and are therefore directly coupled to the lattice by crystalline fields. We calculate the magnitude of T_1 for the two phonon process described above by using as an approximate estimate of the orbit-lattice interaction the Hamiltonian $H_{OL} = (V_2^1 + V_4^1)\epsilon$, where ϵ is the strain and V_2^1 and V_4^1

are terms in the crystalline field potential as defined by Stevens [8]. We find, apart from spin-spin effects,

$$\frac{1}{T_1} \approx \frac{3}{2\hbar\rho v^5} \left| \left\langle \text{ground doublet} \left| V_2^1 + V_4^1 \right| \text{excited state} \right\rangle \right|^2 \left(\frac{\Delta}{\hbar}\right)^3 e^{-\Delta/kT} ,$$

where ρ is the density and v is the phonon velocity. Estimating V_2^1 and V_4^1 from the work of Judd [9] and letting $\Delta/k = 34°K$,

$$T_1 \sim 10^{-10}\, e^{34/T} \text{ secs.} ,$$

in very satisfactory agreement with experiment. It is to be pointed out that at higher temperatures the usual Raman processes may become more important than our two stage process because of virtual transitions to and from the second excited state for which $\Delta/k \sim 200°$ K [4]. For $T < 1.8°K$ the normal direct process would appear to become more important. Over a limited temperature range, however, we find the presence of the low lying excited state to be all important and to give rise to the dominant relaxation mechanism.

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