

**Zeitschrift:** Archives des sciences [1948-1980]  
**Herausgeber:** Société de Physique et d'Histoire Naturelle de Genève  
**Band:** 13 (1960)  
**Heft:** 9: Colloque Ampère

**Artikel:** Spin-lattice relaxation in cerium magnesium nitrate at helium temperatures : a new process  
**Autor:** Finn, C.B.P. / Orbach, R. / Wolf, W.P.  
**DOI:** <https://doi.org/10.5169/seals-738553>

### **Nutzungsbedingungen**

Die ETH-Bibliothek ist die Anbieterin der digitalisierten Zeitschriften auf E-Periodica. Sie besitzt keine Urheberrechte an den Zeitschriften und ist nicht verantwortlich für deren Inhalte. Die Rechte liegen in der Regel bei den Herausgebern beziehungsweise den externen Rechteinhabern. Das Veröffentlichen von Bildern in Print- und Online-Publikationen sowie auf Social Media-Kanälen oder Webseiten ist nur mit vorheriger Genehmigung der Rechteinhaber erlaubt. [Mehr erfahren](#)

### **Conditions d'utilisation**

L'ETH Library est le fournisseur des revues numérisées. Elle ne détient aucun droit d'auteur sur les revues et n'est pas responsable de leur contenu. En règle générale, les droits sont détenus par les éditeurs ou les détenteurs de droits externes. La reproduction d'images dans des publications imprimées ou en ligne ainsi que sur des canaux de médias sociaux ou des sites web n'est autorisée qu'avec l'accord préalable des détenteurs des droits. [En savoir plus](#)

### **Terms of use**

The ETH Library is the provider of the digitised journals. It does not own any copyrights to the journals and is not responsible for their content. The rights usually lie with the publishers or the external rights holders. Publishing images in print and online publications, as well as on social media channels or websites, is only permitted with the prior consent of the rights holders. [Find out more](#)

**Download PDF:** 11.08.2025

**ETH-Bibliothek Zürich, E-Periodica, <https://www.e-periodica.ch>**

## 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  $\text{Ce}_2\text{Mg}_3(\text{NO}_3)_{12} \cdot 24\text{H}_2\text{O}$ , 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 ( $\Delta/k = 34^\circ\text{K}$ ) 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

\* Supported in part by the U.S. Air Force Cambridge Research Center through the European Office of A.R.D.C.

\*\* National Science Foundation Postdoctoral Fellow.

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  $34 \pm 1^\circ\text{K}$ , in good agreement with the value  $\Delta/k \cong 34^\circ\text{K}$  deduced from specific heat and susceptibility.

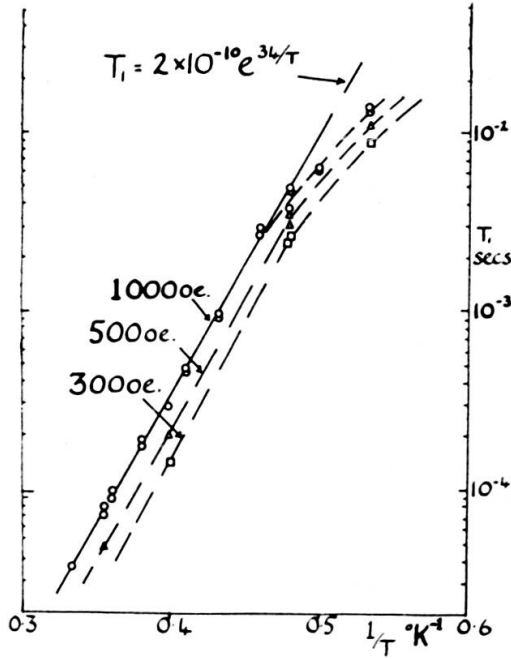


Fig. 1.

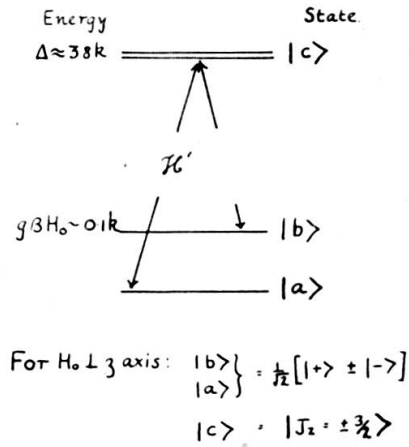


Fig. 2.

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  $\epsilon$  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  $\rho$  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  $\Delta/k = 34^\circ\text{K}$ ,

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

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  $\Delta/k \sim 200^\circ\text{K}$  [4]. For  $T < 1.8^\circ\text{K}$  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.

We wish to thank Dr. R. J. Elliott for many helpful suggestions and Professor J. H. Van Vleck and Dr. A. H. Cooke for stimulating discussions.

#### REFERENCES

1. CASIMIR, H. G. B. and F. K. DU PRÉ, *Physica*, 5, 507 (1938).  
CASIMIR, H. G. B., D. BIJL and F. K. DU PRÉ, *Physica*, 8, 449 (1941).  
BENZIE, R. J. and A. H. COOKE, *Proc. Phys. Soc.*, A 63, 201 (1950).
2. COOKE, A. H., H. J. DUFFUS and W. P. WOLF, *Phil. Mag.*, 44, 623 (1953).
3. C. A. BAILEY, to be published.
4. LEASK, M. J. M. and W. P. WOLF, to be published.
5. VAN VLECK, J. H., *Phys. Rev.*, 57, 426 (1940).
6. Low Temperature Specific Heat Measurements (C. A. BAILEY, *Phil. Mag.*, 4, 833, 1959) indicate that  $\theta_D \sim 60^\circ\text{K}$ .
7. It has been pointed out by Dr. B. Bölger (private communication) that a similar process has been put forward by J. P. LLOYD and G. E. PAKE, *Phys. Rev.*, 94, 579 (1954) in quite a different context. We are indebted to Dr. Bölger for this communication.
8. STEVENS, K. W. H., *Proc. Phys. Soc.*, A 65, 209 (1952).
9. JUDD, B. R., *Proc. Roy. Soc.*, 232, 458 (1955).