

## Electron-Spin-Resonance Linewidth Variation with Temperature in Some Rare-Earth Salts: $T_1/T_2$ Ratios\*

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We have measured the temperature dependence of the electron-spin-resonance linewidth of Ce, Nd, and Sm in lanthanum magnesium nitrate from below 4°K to as high as 26°K. Both Ce and Nd displayed a linewidth which was essentially Lorentzian at the lowest temperature and which broadened with increasing temperature in accordance with an Orbach relaxation mechanism. A comparison of the strength of this broadening with standard relaxation measurements below 4°K indicated that  $T_1/T_2$  was 8.6 for the Nd sample and was estimated to be 13 for the Ce crystal. Sm displayed a temperature-dependent broadening which could be interpreted as due either to a Raman or an Orbach mechanism.

### I. INTRODUCTION

WE have measured the electron-spin-resonance (ESR) linewidth of trivalent cerium, neodymium, and samarium in single crystals of lanthanum magnesium nitrate (LMN) from below 4°K to as high as 26°K. We had anticipated demonstrating the usefulness of this technique for supplementing relaxation measurements made below 4°K in order to determine the strength of the Raman relaxation rate, which is generally far weaker below 4°K than an Orbach process. We found, however, that the linewidth was completely dominated by an Orbach process with a strength that was greater than the extrapolated low-temperature relaxation measurements would predict. This discrepancy was verified for our specific LMN:Nd sample by directly measuring the Orbach relaxation rate between 2.54 and 3.70°K using the pulse-saturation technique and comparing the relaxation data with the linewidth measurements between 9.3 and 26.3°K.

The apparatus, measurement procedures, and results are discussed in Sec. II. In Sec. III we briefly summarize our experimental results and cite a reference which treats the theoretical aspects of this phenomenon.

### II. EXPERIMENTAL

#### A. Apparatus and Procedure

The ESR linewidth measurements were made with a standard ESR X-band reflection spectrometer which was frequency stabilized to the sample cavity and employed straight crystal detection. The magnetic field was modulated at audio frequencies, and the resonance lines were phase-detected so that the peak-to-peak derivative linewidth of the absorption curve was recorded at each temperature. In general, the line was swept through twice at each temperature, once in each

direction, and only with sufficient depth to determine the derivative peaks. Exceptions were those cases where a line shape measurement was made, and then the sweep was essentially through the entire line.

Our cryostat (built by Janis Research Co., Inc.) featured a throttling valve through which liquid helium at 4°K could flow under gravity into a diffuser mounted in the bottom of the insulated inner Dewar. The liquid was converted to gaseous helium by the diffuser and then flowed past the microwave cavity. The sample as well as the germanium thermometers were kept in thermal contact with the walls of the copper cavity by means of apiezon *N* grease. The temperature of the cavity was stabilized with a bucking heater which was regulated by a Cryogenic Research model TC-101 temperature controller using a Minneapolis-Honeywell germanium resistor as a sensing element. The temperature was monitored using a similar, calibrated germanium resistor in one arm of a dc bridge. Resistance values,  $R$ , measured in this way were corrected for the magnetoresistance effect according to the relation<sup>1</sup>  $\Delta R/RH^2 = \alpha$ , where  $\alpha$  was found to be  $2.4 \times 10^{-10}$  Oe<sup>-2</sup> near 4°K and was assumed to be temperature-independent. Microwave power incident on the cavity was typically under 5 mW.

The relaxation measurements on the LMN:Nd sample were made with a superheterodyne pulse spectrometer constructed by Young.<sup>2</sup> The temperatures were measured using a calibrated 250-Hz audio bridge with a Cryocal, Inc. germanium sensor in one arm. Resistance values were corrected for a temperature-dependent magnetoresistance effect,  $\alpha(T)$ .

There is a temperature-independent contribution,  $\Delta H_0$ , to the peak-to-peak derivative linewidth,  $\Delta H$ ;  $\Delta H_0$  is typically a few Oe for rare-earth-doped LMN.

<sup>1</sup> Frank L. Blatt, *Physics of Electronic Conduction in Solids* (McGraw-Hill Book Co., New York, 1968), Chap. 8, p. 287.

<sup>2</sup> B. A. Young, Ph.D. thesis, University of Illinois, 1967 (unpublished).

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At low concentrations (under 0.1 at.%) and low temperatures (below 4°K), the line shape in these salts has been found to be approximately Gaussian<sup>3</sup> with a very anisotropic width, apparently due to random local strains. At higher concentrations, dipolar effects should produce a Lorentzian line at these low temperatures, and at higher temperatures, short relaxation times should produce a Lorentzian line shape according to the Bloch equations.<sup>4</sup> The relationship between the transverse relaxation time  $T_2$  and the associated peak-to-peak derivative linewidth  $\Delta H'$  follows from the Bloch equations and is

$$1/T_2 = 7.62 \times 10^6 g \Delta H' \text{ sec}^{-1}, \quad (1)$$

where  $\Delta H'$  is in Oe and  $g$  is the spectroscopic splitting factor for the magnetic doublet. In the temperature range where the ESR lines are broadened by the very short relaxation times ( $T_1$ ), one might reasonably expect the ratio  $T_1/T_2$  to be a constant which is close, if not identical, to unity.

The relationship between  $\Delta H'$ ,  $\Delta H_0$ , and the observed peak-to-peak derivative linewidth  $\Delta H$  depends upon the line shapes associated with  $\Delta H'$  and  $\Delta H_0$ . For two folded Gaussian line shapes

$$\Delta H' = (\Delta H^2 - \Delta H_0^2)^{1/2} \quad (\text{Gaussian}) \quad (2)$$

and for two folded Lorentzian line shapes

$$\Delta H' = \Delta H - \Delta H_0 \quad (\text{Lorentzian}), \quad (3)$$

while no simple expression exists for a mixture of the two shapes.<sup>5</sup> For most of the data both corrections were very small, and specific details will be discussed later.

The corresponding values of  $\Delta H'$  and temperature were fitted to a general expression of the form

$$\Delta H' = B' / [\exp(\Delta/T) - 1] + C'T^n, \quad (4)$$

where  $B'$ ,  $\Delta$ ,  $C'$ , and  $n$  could be fixed or varied in order to obtain a least-squares fit. The first term on the right in Eq. (4) corresponds to an Orbach relaxation process.<sup>6</sup> The last term in Eq. (4) represents a two-parameter approximation to a Raman relaxation process. The usual expression<sup>7</sup> of  $T^9$  or  $T^9 J_8(\theta/T)$  for the temperature dependence of the Raman process has not been successful in describing some rare-earth relaxation data.<sup>8</sup> Kiel and Mims propose a more precise formulation<sup>8</sup> of the Raman process but the results cannot be expressed analytically. Instead, they are able to fit their Raman relaxation data to a  $C'T^n$  law and obtain  $n$  values between 10 and 11. Huang<sup>9</sup> has considered

the effects of optical phonons and obtained two-phonon relaxation rates which are proportional to  $\exp(-\hbar\omega_0/kT)$  at low temperatures. Here  $\omega_0$  is an optical phonon frequency at  $k=0$ . No term representing a direct process has been included in Eq. (4) since it would be negligible in the region of interest where  $\Delta H' \geq \Delta H_0$ .

## B. Results

Figure 1 shows the results of linewidth measurements on LMN:Ce. A chemical analysis indicated the actual Ce content to be 0.9 at.%. In this hexagonal lattice, Ce has principal  $g$  values of 1.83 and 0.02 in the perpendicular and parallel orientations, respectively.<sup>3,10,11</sup> The measurements were made for a perpendicular orientation. This sample was warmed to room temperature between periods of data taking and probably as a result of this cycling was strained sufficiently to change the ESR line at 4°K from an asymmetric singlet to a barely resolvable doublet. As would be expected, the distortion in both cases was on the high-field side of the resonance. An analysis of the low-field side of both the singlet and the lower doublet showed them to be Lorentzian, characterized by a full peak-to-peak derivative width of  $10.3 \pm 2.3$  Oe which we took as the value of  $\Delta H_0$ . Because of the increase in the linewidth as the temperature is raised, our measurements at 6.34 and 9.29°K showed the ESR line to be

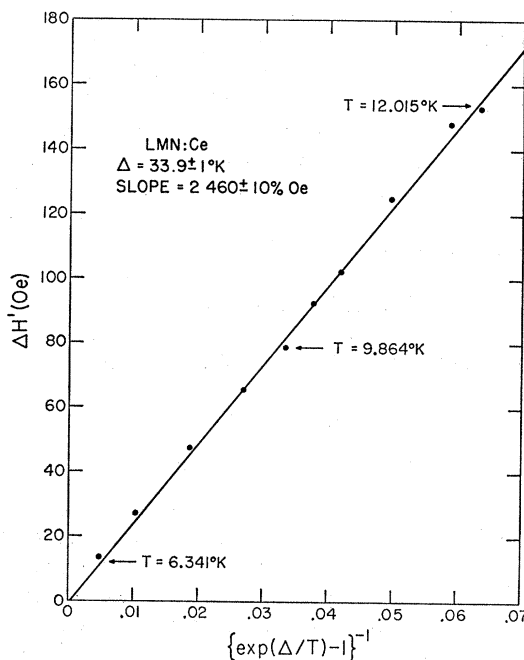


Fig. 1. Relaxation-induced peak-to-peak derivative linewidths  $\Delta H'$  of 0.9 at.% Ce in lanthanum magnesium nitrate plotted against the phonon occupation number at energy  $\Delta$ . These data indicate a dominant Orbach relaxation process.

<sup>3</sup> P. L. Scott, H. J. Stapleton, and C. Wainstein, *Phys. Rev.* **137**, A71 (1965).

<sup>4</sup> F. Bloch, *Phys. Rev.* **70**, 460 (1946).

<sup>5</sup> D. W. Posener, *Australian J. Phys.* **12**, 184 (1959).

<sup>6</sup> R. Orbach, *Proc. Roy. Soc. (London)* **A264**, 458 (1961).

<sup>7</sup> J. H. Van Vleck, *Phys. Rev.* **57**, 426 (1940).

<sup>8</sup> A. Kiel and W. B. Mims, *Phys. Rev.* **161**, 386 (1967).

<sup>9</sup> Chao-Yuan Huang, *Phys. Rev.* **154**, 215 (1967).

<sup>10</sup> M. J. M. Leask, R. Orbach, M. J. D. Powell, and W. P. Wolf, *Proc. Roy. Soc. (London)* **A272**, 371 (1963).

<sup>11</sup> K. H. Langley and C. D. Jeffries, *Phys. Rev.* **152**, 358 (1966).

symmetric and Lorentzian with no evidence of a twin or cracked crystal. Because of these factors, Eq. (2) was used to obtain  $\Delta H'$ . Figure 1 indicates that these values of  $\Delta H'$  can be fitted very well with only an Orbach process. The average absolute deviation in Fig. 1 is only 2.1 Oe and the correlation coefficient is 0.9986. Furthermore, the best fitting value of the parameter  $\Delta$  is 33.9°K, which is in good agreement with the  $T_1$  measurements on LMN:Ce reported by five other investigators and summarized by Ruby *et al.*<sup>12</sup> as

$$1/T_1 = B \exp(-\Delta/T) \quad (5)$$

with  $B = 2.7 \times 10^9 \text{ sec}^{-1}$  and  $\Delta = 34^\circ\text{K}$ . Brya and Wagner<sup>13</sup> have fitted their relaxation data on LMN:Ce between 1.25 and 2.75°K to a direct process, an Orbach process with a  $\Delta$  value of 36.7°K and a Raman rate at  $T^9$ . This value of  $\Delta$  is in closer agreement with an infrared determination<sup>14</sup> of  $36.25 \pm 0.4^\circ\text{K}$ . A modification of our fit to include a term  $C'T^9$  yielded a negligible change and a value of  $[(7 \pm 20) \times 10^{-10}]^\circ\text{K}^{-9} \text{ Oe}$  for  $C'$ . The best fitting value of  $B'$  is 2460 Oe with an estimated error of 10%. By means of Eqs. (1), (4), and (5) with  $B = 2.7 \times 10^9 \text{ sec}^{-1}$  from the relaxation data and  $B' = 2460 \text{ Oe}$  from our linewidth data, one calculates the ratio of  $T_1/T_2$  to be 13. Unfortunately the fact that our sample was cracked prevented us from making meaningful pulse saturation measurements of  $T_1$ . It is worth noting that some unpublished data of Culvahouse and Addis<sup>15</sup> on the temperature dependence of  $\Delta H'$  in 100% CeMN can be fitted to Eq. (4) with  $B' = 5350 \text{ Oe}$ ,  $\Delta = 34^\circ\text{K}$ , and  $C' = 0$ .

The LMN:Nd crystal was grown at ice temperature from a solution containing 1 at.% Nd. No chemical analysis of the actual sample was made. The principal  $g$  values in the perpendicular and parallel directions are 2.71 and 0.368,<sup>3,16</sup> respectively. Our measurements were made for a perpendicular orientation. The line-shape of the resonance was observed near 4, 6, 10, 12, and 14°K and found to be Lorentzian and the low-temperature measurements yielded a value of 3.3 Oe for  $\Delta H_0$ . The best fit of the raw data to an Orbach relaxation term using Eq. (3) (Lorentzian line shapes) yielded the parameters  $B' = 1099 \pm 50 \text{ Oe}$  and  $\Delta = 49.1 \pm 0.9^\circ\text{K}$ ; while the best fit of the same raw data to the same process using Eq. (2) (Gaussian line shapes) yields parameters  $B' = 980 \pm 44 \text{ Oe}$  and  $\Delta = (46.3 \pm 0.8)^\circ\text{K}$ . The Gaussian type of correction yields a value of  $\Delta$  in better agreement with the relaxation measurements, discussed below, and with the value of 47.6°K deduced from optical studies<sup>17</sup> on 100% NdMN. Theory and

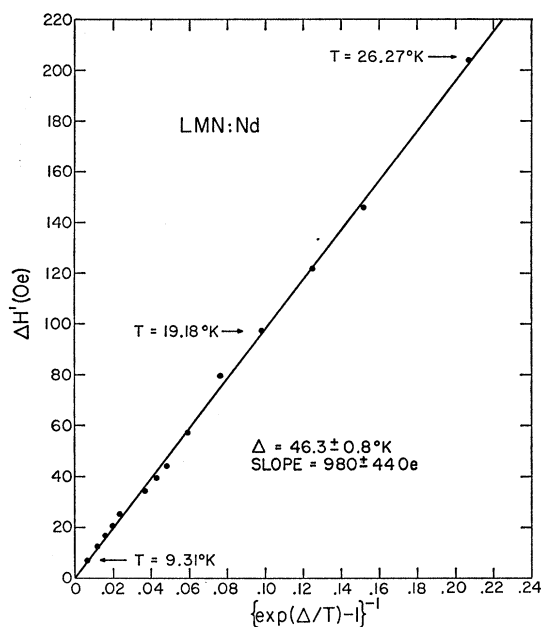


FIG. 2. Relaxation-induced peak-to-peak derivative linewidths  $\Delta H'$  of Nd in lanthanum magnesium nitrate plotted against the phonon occupation number at energy  $\Delta$ . These data indicate a dominant Orbach relaxation process.

experiments<sup>2,18,19</sup> show that values of  $\Delta$  deduced from relaxation measurements according to the second term in Eq. (4) may be less than values determined optically but that any discrepancy diminishes as the temperature is raised. For these reasons Fig. 2 displays data corrected with Eq. (2) for Gaussian line shapes. These values of  $\Delta H'$  can be closely fitted with only an Orbach relaxation term. The average absolute deviation is only 1.7 Oe and the correlation coefficient is 0.99934. If a term involving  $C'T^9$  is added to the fit, a value of  $[(6 \pm 10) \times 10^{-11}]^\circ\text{K}^{-9} \text{ Oe}$  results.

Figure 3 shows the results of directly measuring the spin-lattice relaxation time  $T_1$  in the same sample by pulse saturation techniques in a temperature region where the Orbach process dominates. The strength of the Orbach relaxation process is  $B = 2.34 \times 10^9 \text{ sec}^{-1}$  with an error of 20% and  $\Delta = (45.3 \pm 0.6)^\circ\text{K}$ . These relaxation data are more scattered than the linewidth data and the correlation coefficient is only 0.9901. If the slight difference in  $\Delta$  values is ignored, then the data of Figs. 2 and 3 imply a  $T_1/T_2$  ratio of 8.6. If the different values are not ignored then the extrapolated  $T_1/T_2$  ratio is still 7.8 at 9°K and 8.3 at 27°K. Furthermore, if the 46.3°K value is assumed to be the true splitting in the dilute salt, then a Gaussian distribution of  $\Delta$  values with a width  $2\sigma = 4^\circ\text{K}$  about this value of 46.3°K could explain the one-degree shift<sup>2,18,19</sup> in the fitted  $\Delta$  of the low-temperature relaxation data. A

<sup>12</sup> R. H. Ruby, H. Benoit, and C. D. Jeffries, Phys. Rev. **127**, 51 (1962).

<sup>13</sup> William J. Brya and Peter E. Wagner, Phys. Rev. **147**, 239 (1966).

<sup>14</sup> J. H. M. Thornley, Phys. Rev. **132**, 1492 (1963).

<sup>15</sup> J. W. Culvahouse (private communication).

<sup>16</sup> A. H. Cooke and H. J. Dufus, Proc. Roy. Soc. (London) **A229**, 407 (1955).

<sup>17</sup> B. R. Judd, Proc. Roy. Soc. (London) **A232**, 458 (1955).

<sup>18</sup> B. A. Young and H. J. Stapleton, Phys. Letters **21**, 498 (1966); Bull. Am. Phys. Soc. **12**, 291 (1967).

<sup>19</sup> B. A. Young and H. J. Stapleton, Phys. Rev. **176**, 502 (1968).

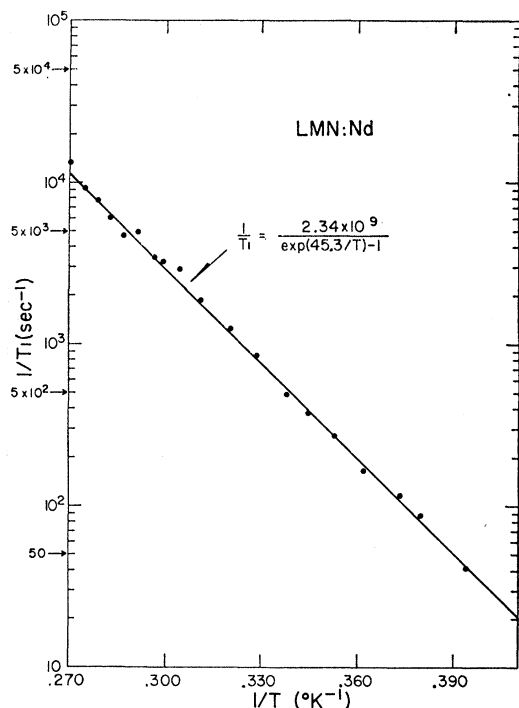


FIG. 3. Relaxation data from pulse-saturation measurements on the same sample as in Fig. 2. These data indicate a dominant Orbach relaxation mechanism and together with the data of Fig. 2 imply a  $T_1/T_2$  ratio of 8.6.

15% discrepancy in the reported values of  $\Delta$  in Ce-doped  $\text{LaCl}_3$  as measured optically and by spin-lattice relaxation can be explained<sup>2,19</sup> by a similar distribution 9  $\text{cm}^{-1}$  wide; a number which is in good agreement with an infrared measurement.<sup>19</sup>

In hopes of observing a clear-cut Raman process, we measured the linewidth of LMN:Sm since the Debye temperature of the host lattice is 61.5°K<sup>20</sup> and the first-excited state in 100% SmMN is 66.9°K.<sup>21</sup> In LMN:Sm the  $\text{Sm}^{3+}$  ion is strongly rejected during growth<sup>22</sup> so the crystals were grown from a 20% Sm-doped solution, and no analysis was made on the resultant crystal. The  $g$  values are 0.363 and 0.736 in the perpendicular and parallel orientation, respectively.<sup>16,22</sup> The linewidths were measured for a parallel orientation with the resonant microwave frequency lowered to about 8.5 GHz. A lineshape analysis showed the line to be essentially Gaussian with a peak-to-peak derivative width of  $2.97 \pm 0.03$  Oe at 3.685°K; intermediate between Gaussian and Lorentzian at 4.032°K with a width of  $2.93 \pm 0.09$  Oe; and Lorentzian at 7.862°K with a width of  $14.94 \pm 0.22$  Oe. The values of  $\Delta H'$  were obtained with Eq. (2) using a value of 2.95 Oe for  $\Delta H_0$ .

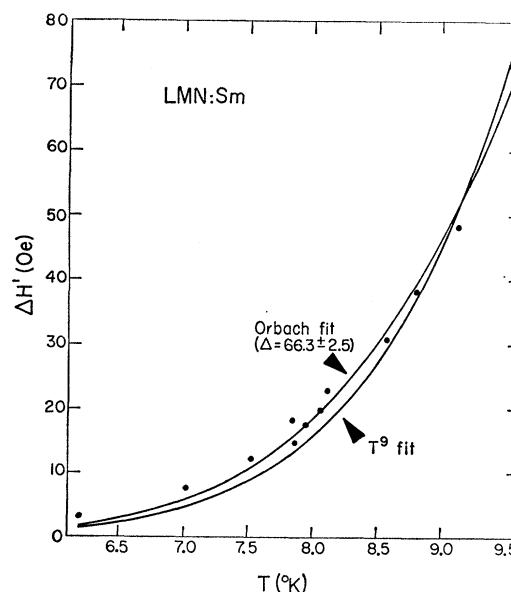


FIG. 4. Relaxation induced peak-to-peak derivative linewidth  $\Delta H'$  of Sm in lanthanum magnesium nitrate plotted against temperature. The best fitting theoretical curves for an Orbach process or a  $T^9$  law are shown. The best fitting power law is  $T^{7.66}$  (not shown), which produces an average absolute deviation of 1.2 Oe, versus 1.7 Oe for the Orbach fit.

The results are shown in Fig. 4 and are ambiguous with respect to the nature of the process. If a pure Orbach mechanism is assumed,  $B' = 7.3 \times 10^4$  Oe with an error of 30% and  $\Delta$  is  $(66.3 \pm 2.5)^\circ\text{K}$ , which is in good agreement with the optical value in 100% SmMN. With such a fit the average absolute deviation is 1.7 Oe, the variance is 4.2  $\text{Oe}^2$ , and the correlation coefficient is 0.9950. On the other hand, if a  $T^9$  law is assumed,  $C'$  is  $(1.17 \pm 0.03) \times 10^{-7} \text{Oe}^{-9}$ . With such a fit the average deviation is 2.7 Oe, the variance is 9.7  $\text{Oe}^2$ , and the correlation coefficient is 0.987. The best fitting power law is  $T^{7.66}$  with an error of  $\pm 0.2$  and a coefficient of  $(2.2 \pm 1.1) \times 10^{-4}$ . The resultant indicators of the fit are an average absolute deviation of 1.2 Oe, a variance of 2.6  $\text{Oe}^2$ , and a correlation coefficient of 0.9973. It does not seem possible to determine the nature of the relaxation process on the basis of this analysis.

### III. DISCUSSION

The results we have presented for Ce and Nd in LMN give strong evidence that associated with an Orbach relaxation ( $1/T_1$ ) process, there can be an even stronger line broadening ( $1/T_2$ ) mechanism so that  $T_1/T_2$  is greater than unity. Our conclusion that we observe only an Orbach process in these two salts is based upon plausibility. It seems most unlikely that in both LMN:Ce and LMN:Nd a dominant Raman relaxation process could follow the temperature dependence of an Orbach mechanism with the correct  $\Delta$  value.

<sup>20</sup> C. A. Bailey, Proc. Phys. Soc. (London) **83**, 369 (1964).

<sup>21</sup> A. Friederich, K. H. Hellwege, and H. Lammermann, Z. Physik **159**, 524 (1960).

<sup>22</sup> P. L. Scott and C. D. Jeffries, Phys. Rev. **127**, 32 (1962).

In the following paper, Culvahouse and Richards<sup>23</sup> examine the relationship between  $T_1$  and  $T_2$  due exclusively to an Orbach relaxation mechanism and conclude that  $T_1$  and  $T_2$  must be equal for the cases we have investigated if the measured Orbach relaxation time  $T_1$  represents the true spin-phonon relaxation

time and not a longer spin-bath relaxation time due to a phonon bottleneck.

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<sup>23</sup> J. W. Culvahouse and Peter M. Richards, following paper, Phys. Rev. **178**, 485 (1969).

## $T_1$ and $T_2$ for Orbach Relaxation Processes\*

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The master equation for the density matrix of the ionic levels involved in the relaxation process described by Orbach is developed in a form valid for low temperatures. In the absence of degeneracies, the equations for the diagonal elements of the density matrix are the rate equations for the populations which have been solved before; and the equations for the off-diagonal elements show that  $(1/T_2)$ , the linewidth for a transition between two levels, is given by one-half the sum of the phonon-induced rates out of those levels. Cases in which the inverse lifetimes of the excited states are not small compared with the splitting of the doublets are investigated, and we find that the modification of  $T_1$  can be quite significant, but that the effect on  $T_2$  is slight in all cases. For  $\text{Ce}^{3+}$ ,  $\text{Nd}^{3+}$ , and  $\text{Sm}^{3+}$  in the double nitrates, we find that  $T_1 = T_2$  within 12.5% for the magnetic field perpendicular to the symmetry axis for all conditions. For  $\text{Ce}^{3+}$  in  $\text{LaMg}$  double nitrate with the field perpendicular to the symmetry axis, measurements at 10 Gc/sec should give  $T_1 = T_2$  within 3%, whereas the experimental result reported by Stapleton and Brower is  $T_1 = 13T_2$ . It is found that spin-spin interactions between the ground doublet and the excited doublet cannot be the cause of the discrepancy, but that a near-universal bottlenecking of a part of the phonon modes would lead to an explanation of the linewidth measurements and not be at variance with the size and concentration dependence of  $T_1$  thus far reported for the double-nitrate crystals. This hypothesis is based on the calculations of Orbach and Vrederoe, which show that the lifetime of the transverse phonon modes may be much longer than that for the longitudinal modes. Further, the transverse phonons should produce the greater part of the orbit-lattice interaction. This hypothesis suggests that a true spin-lattice  $T_1$  would be measured only at very low concentrations, and that with increasing concentrations a bottleneck would develop so that the apparent relaxation time would increase with concentration until the bottlenecked relaxation became comparable with the rate due to other unbottlenecked phonons acting in parallel with the bottle-necked phonons. The experiments of Adde and Geschwind appear to show some aspects of this behavior.

### I. INTRODUCTION

THE measurements of Stapleton and Brower<sup>1</sup> reported in the preceding paper demonstrate a temperature-dependent EPR linewidth for the lowest doublet of  $\text{Ce}^{3+}$  and  $\text{Nd}^{3+}$  in double-nitrate crystals which strongly suggests that the width arises from a transverse relaxation time  $T_2$  associated with the Orbach relaxation process. If this is so, then  $T_2$  is an order of magnitude shorter than  $T_1$ .

There is other, less extensive, evidence that  $T_2$  is considerably less than  $T_1$  for Orbach relaxation processes in rare-earth ions. The early pulse work of

Cowan and Kaplan<sup>2</sup> with  $\text{Ce}^{3+}$  in the double nitrates suggests conclusions in accord with Stapleton and Brower. The temperature-dependent  $T_2$  which their data show could be very well explained as a temperature-independent component plus a term  $\frac{1}{10} \times T_1$ . Mims<sup>3</sup> has also noted that the phase memory time,  $T_M$  as he denotes it, which should be the true  $T_2$ , is an order of magnitude shorter than  $T_1$  in the high-temperature regions for Ce and Er in  $\text{CaWO}_4$ , where it is probable that an Orbach mechanism dominates the longitudinal relaxation.

In this paper we investigate  $T_1$  and  $T_2$  in the Orbach process and conclude that they are equal in the cases investigated by Stapleton and Brower if the  $T_1$  is not affected by phonon heating (bottlenecking).<sup>4</sup> We suggest

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<sup>1</sup>H. J. Stapleton and K. L. Brower, preceding paper, Phys. Rev. **178**, 481 (1969).

<sup>2</sup>J. A. Cowan and D. E. Kaplan, Phys. Rev. **124**, 1098 (1962).

<sup>3</sup>W. B. Mims, Phys. Rev. **168**, 370 (1968).

<sup>4</sup>J. H. Van Vleck, Phys. Rev. **59**, 724 (1941).