between a severe phonon bottleneck and the condition for the build-up of stimulated phonon emission, i.e., maser oscillations. In our notation this condition is, from Eq. (40c),

$$-n/n_0 \geq \sigma^{-1} \tag{75}$$

so that only a fraction $\sigma^{-1} \approx 10^{-3}$ of the spins need inverting; such experiments are in progress here.

ACKNOWLEDGMENTS

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Paramagnetic Resonance below 1°K: Spin-Lattice Relaxation of Ce^{3+} and Nd^{3+} in Lanthanum Magnesium Nitrate*

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An apparatus is described for performing paramagnetic resonance experiments at $\nu \approx 9.6$ kMc/sec at temperatures down to $T \approx 0.2$°K, obtained by adiabatic demagnetization. It is used to extend the spin-lattice relaxation rate T_1^{-1} measurements of the previous paper on Nd in the double nitrate $La_2Mg_3(NO_3)_{12} \cdot 24H_2O$ down to 0.3°K, where the direct process is very well displayed. For $z \perp H$ the result is $T_1^{-1} = 0.3 \coth(h\nu/2kT)$ sec^{-1}, to be compared to the theoretical estimate $T_1^{-1} = 0.6 \coth(h\nu/2kT)$ sec^{-1}. At the lowest temperatures the relaxation rate tends toward the temperature-independent value of spontaneous phonon emission.

For Ce in the double nitrate, the observed spin-bath relaxation rate T_b^{-1} at temperatures below 1°K is not the direct spin-lattice relaxation rate, but rather a phonon-limited bottleneck rate $T_b^{-1} \propto \coth^2(h\nu/2kT)$ which is concentration dependent. The results are consistent with the bottleneck rate $T_b^{-1} \propto T^2$ observed in the previous paper for other ions at higher temperatures. We find a lower limit for the true direct process $T_1^{-1} > 20 \coth(h\nu/2kT)$ sec^{-1}.

I. INTRODUCTION

ALTHOUGH microwave paramagnetic resonance experiments at temperatures T below 1°K have not yet been reported in detail, they are not without interest. Some phenomena which come to mind are: (1) The direct process for the spin-lattice relaxation rate T_1^{-1} in paramagnetic salts should become dominant over two-phonon processes and, if $h\nu \gg kT$, T_1^{-1} should become temperature independent, corresponding to spontaneous emission of phonons. (2) The phonon bottleneck may become appreciable.[1] (3) The Van Vleck linewidth of a resonance line should decrease as the spins become aligned at low temperatures,[2] and the line may become resolved into several sharp lines. (4) Spin ordering may set in due to dipolar interaction, or to exchange interactions even in magnetically dilute systems. (5) Starting temperatures $T \sim 0.2$°K are useful in dynamic nuclear cooling experiments.[3]

To investigate these effects we have constructed a paramagnetic resonance spectrometer operating at a frequency $\nu \approx 9.6$ kMc/sec and temperatures down to $T \approx 0.25$°K. In this paper we describe experiments concerning topics (1) and (2) above. They have been briefly reported earlier,[4] and are, to some extent, low-temperature extensions of the work described in the preceding paper[1] (referred to here as SJ), whose notation we use throughout.

II. APPARATUS

The essential details of the low-temperature paramagnetic resonance apparatus are given in Fig. 1. A long stainless steel evacuated can (shown cut-away) contains a cooling salt pill thermally linked to a microwave cavity. With the magnet in position A and the mechanical heat switch closed, the pill is cooled to $T_i \approx 1.5$°K by the pumped He^4 bath in a field $H_i \approx 9$ kOe. The switch is opened and the magnet lowered to position B, whereby the pill is cooled by adiabatic demagnetization to $T_f \approx 0.25$°K for the salt used at present. Within a minute or so the cavity is also cooled to this temperature. The system then slowly warms up to 1.5°K over a period of 4 to 6 h, during which paramagnetic resonance experiments are performed on

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* Supported in part by the Atomic Energy Commission and the Office of Naval Research.

† NATO Fellow.

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[3] C. Kittel, Physica (Suppl.) **24**, 588 (1958).

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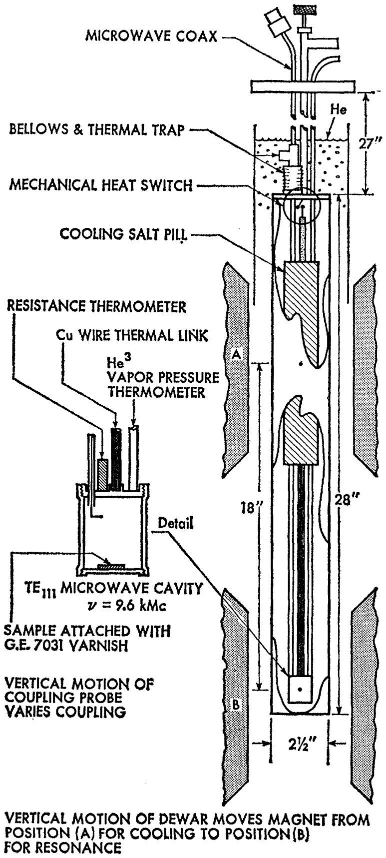

FIG. 1. Paramagnetic resonance apparatus used for operation at temperatures down to $T \approx 0.25$°K. The liquid nitrogen jacket is not shown.

samples contained in the cavity in fields up to 9 kOe. The fringe field on the salt pill is negligible and the cavity temperature is quite independent of the applied field.

The present salt pill consists of 150 grams of powdered $\mathrm{Mn(NH_4)_2(SO_4)_2 \cdot 6H_2O}$, mixed with toluene and G.E. 7031 varnish, contained in a thin brass can in which are many copper wire fingers. The electromagnet has 12-in. diameter poles and a $2\frac{5}{8}$-in. gap, and has a highly stabilized current supply. The temperature T of the cavity is measured by a He^3 vapor pressure thermometer, which is used to calibrate, in the range $0.45° \leqq T \leqq 1.8$°K, a Speer Company 100-ohm carbon resistor as a secondary thermometer. The resistance R was fitted to the empirical relation

$$\log_{10}R + (a/\log_{10}R) = (b/T) + c,$$

which was then extrapolated to the lowest temperatures used.

The microwave cavity is a thin-walled, gold-plated, brass cylinder operating in the TE_{111} mode, which produces a field H_1 tangent to the removable bottom cap, upon which the crystal sample is mounted with G.E. 7031 varnish. The cavity is coupled to a miniature coaxial transmission line made of Teflon-clad bronze wire (Microdot Inc., cable No. 93-3908) inserted into a stainless steel tube. A quarter-wave short is used as a thermal trap at 1.5°K.

The experiments reported here consist in the measurement of the spin-bath relaxation rate T_b^{-1} for Nd^{3+} and for Ce^{3+} in single crystals of $\mathrm{La_2Mg_3(NO_3)_{12} \cdot 24H_2O}$ ("La*MN*") by the method described in SJ. Figure 2 is a block diagram of the spectrometer, which is quite similar to that of SJ, the essential difference being that it is a reflection system using a magic tee. The paramagnetic resonance was partially saturated by a pulse of microwave power using a diode switch; the transient recovery of the signal at a monitoring power was then photographed from an oscilloscope connected to the video output of a microwave superheterodyne receiver. In order to avoid saturation and sample heating, the monitor power level was usually set at about 10^{-10} W. The photographed decays were replotted on semilog paper to determine if the decay was exponential and, if so, the time constant T_b. After a short nonexponential behavior, the decay usually became exponential, and the time constant T_b was measured in the tail of the curve where the spins had almost cooled to the bath temperature. It was observed, however, that long, strong pulses heated the sample and cavity appreciably, and a nonexponential thermal recovery time of several seconds could be observed both by the cavity resistance thermometer and the resonance signal. This effect was clearly distinguishable from the spin-bath relaxation time T_b, which was found to be independent of pulse repetition rate, power, and duration for sufficiently weak pulses.

III. SPIN-LATTICE RELAXATION OF Nd^{3+} IN $\mathrm{La_2gM_3(NO_3)_{12} \cdot 24H_2O}$

Measurements of T_b for cavity temperatures between 0.3° and 2°K were made for two crystals of La*MN*, one containing 5% Nd of natural isotopic abundance and the other 1% Nd enriched to 98.5% even isotopes. The frequency used was $\nu = 9.67$ kMc/sec, correspond-

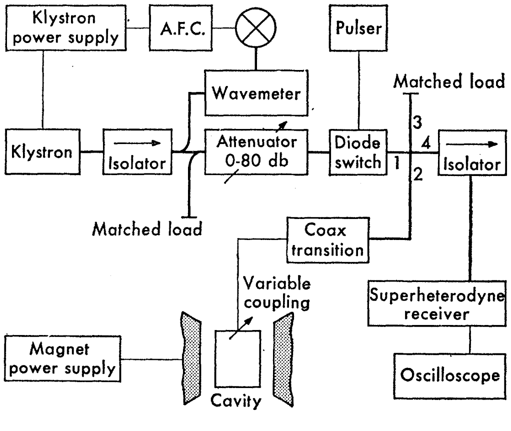

FIG. 2. Block diagram of the microwave apparatus used to measure spin-bath relaxation times.

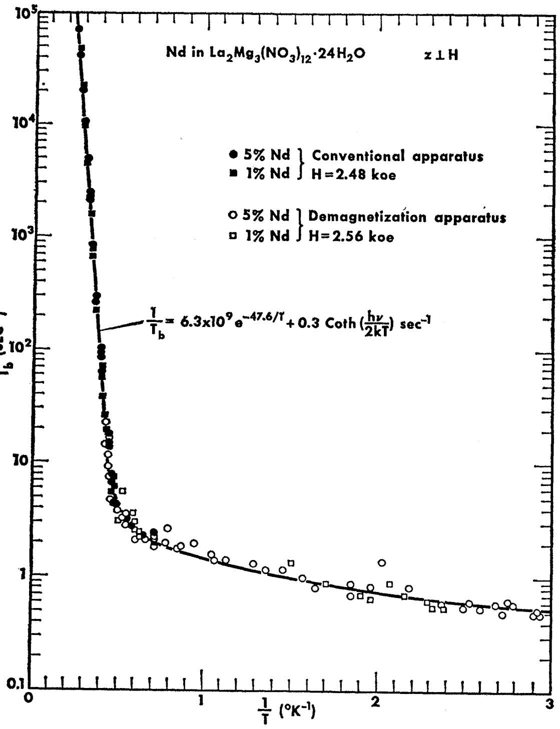

FIG. 3. Experimental results for the spin-bath relaxation rate T_b^{-1} for Nd in single crystals in LaMN in the temperature range $0.3<T<4.3$°K. The data for the higher temperatures (solid squares and circles) are taken from the preceding paper (SJ, Fig. 5). The over-all data are explained by an Orbach process plus a direct process. At the lowest temperatures the relaxation rate is chiefly due to spontaneous emission of phonons.

ing to a resonance field $H=2.56$ kOe for the strong central $(I=0)$ line of the Nd^{3+} spectrum with $z\perp H$. The results are shown in Fig. 3, along with those of SJ (Fig. 5) for the same two crystals in the range 1.5 to 4.2°K. We note that the two sets of data are consistent where they overlap, and that T_b^{-1} is independent of Nd concentration. The over-all data fit quite well the expression

$$T_b^{-1}=6.3\times10^9 \exp(-47.6/T) +0.3 \coth(h\nu/2kT) \text{ sec}^{-1}. \quad (1)$$

Again, the first term represents the Orbach process T_{1O}^{-1}. The second term represents the direct process T_{1d}^{-1}, which from SJ [Eq. (6)] should have the exact form (valid for $h\nu\gg kT$):

$$T_{1d}^{-1}=K \coth(h\nu/2kT) \text{ sec}^{-1}, \quad (2)$$

where $K=A'(h\nu/2k)$ and A' is explicitly defined in SJ [Eq. (18)] and theoretically estimated [Eq. (49)] to be $A'\approx2.6$ sec^{-1} deg^{-1}. We thus predict

$$T_{1d}^{-1}\approx0.6 \coth(h\nu/2kT) \text{ sec}^{-1}, \quad (3)$$

in reasonable agreement with the measured rate T_b^{-1}, which, in the absence of a phonon bottleneck, represents T_{1d}^{-1}.

In particular, we note that the temperature dependence $\coth(h\nu/2kT)$ is very well displayed by the data. As T becomes small compared to $h\nu/k$, the direct relaxation rate tends toward the temperature-independent value $T_{1d}^{-1}=K$, which is just the transition probability for spontaneous emission of phonons. It is easily seen from the rate equation [SJ, Eq. (4)],

$$\dot{N}_b=K[-N_b(\bar{p}_0+1)+N_a\bar{p}_0], \quad (4)$$

that as $T\rightarrow0$, $\bar{p}_0\rightarrow0$, leaving only the spontaneous emission term $\dot{N}_b=-KN_b$. Thus, from our data we are able to say that for Nd^{3+} ions in LaMN with $z\perp H$ in a field $H=2.56$ kOe the direct relaxation time would be $T_{1d}=K^{-1}\cong3.3$ sec at absolute zero.

IV. SPIN-BATH RELAXATION OF Ce^{3+} IN La$_2$Mg$_3$(NO$_3$)$_{12}$·24H$_2$O

The spin-lattice relaxation of Ce^{3+} in LaMN in the liquid He^4 temperature range has been shown to display the Orbach process, first by Finn, Orbach, and Wolf[5] using the Casimir-du Pré method, and also by Leifson and Jeffries,[6] and Scott[7] using the microwave transient method of this paper, and by Cowen and Kaplan[8] using a microwave spin-echo method. All these results are shown in Fig. 4 and are seen to be in substantial agreement; the over-all data fit best the expression

$$T_b^{-1}=2.7\times10^9 \exp(-34/T) \text{ sec}^{-1} \quad (5)$$

in the Orbach region; there is a suggestion of the direct process beginning at $T<1.5$°K.

In order to investigate the direct process in detail we have made measurements of T_b^{-1} in the temperature range 1.8 down to 0.25°K on two crystals of LaMN, the first containing 2% Ce, the second 0.2% Ce. For $\nu=9.67$ kMc/sec, $H=3.80$ kOe and $z\perp H$ our results are given in Fig. 4 and are consistent with the earlier data at the higher temperatures. In our main region of interest, below 1°K, the relaxation rate is concentration dependent and fits the expressions

$$T_b^{-1}=0.8 \coth^2(h\nu/2kT) \text{ sec}^{-1}, \quad \text{for } 2\% \text{ Ce}, \quad (6a)$$

$$T_b^{-1}=2.4 \coth^2(h\nu/2kT) \text{ sec}^{-1}, \quad \text{for } 0.2\% \text{ Ce}. \quad (6b)$$

This is clearly not the direct process, Eq. (2), but rather the phonon bottleneck process given by SJ [Eq. (38a)]:

$$\frac{1}{\tau_1}=\frac{1}{T_{\text{ph}}}\,\frac{12\pi\nu^2\Delta\nu}{v^3c} \coth^2(h\nu/2kT) \text{ sec}^{-1}, \quad (7)$$

where c is the concentration of Ce ions, $\Delta\nu$ is the resonance linewidth, and T_{ph} is the phonon-bath relaxation time. We consider that the bath in the present case is the microwave cavity at temperature T.

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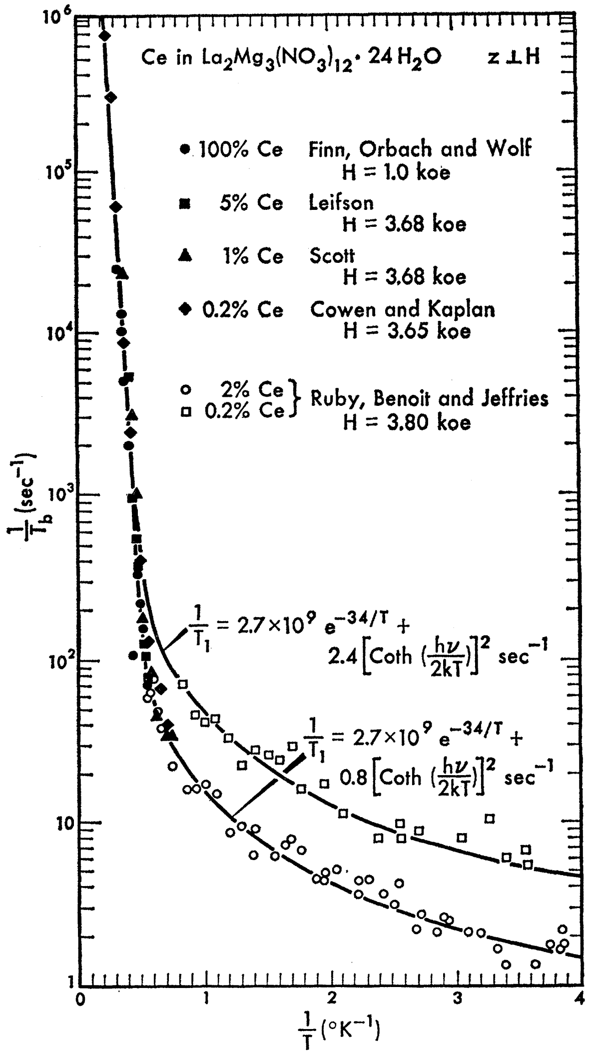

FIG. 4. Experimental values of the spin-bath relaxation rate T_b^{-1} for Ce in single crystals of LaMN. The solid points represent data by other workers in the liquid helium temperature range. The open circles and squares represent the data of this paper at temperatures down to $T\approx 0.25$°K. The over-all data are explained by an Orbach process plus a direct process obscured by a phonon bottleneck, which yields a $\coth^2(h\nu/2kT)$ temperature dependence.

Equation (7) was derived by solving for the approximate time constants of the linearized spin-phonon rate equations [SJ, Eqs. (37a,b)] in the case of a severe bottleneck, i.e., for the case where the spin-phonon coupling is much stronger than the phonon-bath coupling. After a disturbance, e.g., a saturating pulse, the spins and phonons quickly come to the same temperature ($T_s\approx T_p$) and together approach the bath temperature T exponentially with a time constant τ_1. We now rederive Eq. (7) by a simple model which provides more physical insight. Consider a flat crystal with faces of area A, and thickness $l\ll\sqrt{A}$, containing N spins, distributed over two magnetic levels, a and b [cf. SJ, Fig. 1]. The observed paramagnetic resonance absorption signal will be proportional to the difference in population $n=N_a-N_b=Nz$, where we define $z=\tanh(h\nu/2kT_s)\approx\tanh(h\nu/2kT_p)$, $z_0=\tanh(h\nu/2kT)$. As the spin temperature T_s cools after the pulse, the decrease in the Zeeman energy in a time dt will be $dE_z=\frac{1}{2}h\nu Ndz$. This energy dE_z must be carried away by the hot phonons, i.e., acoustic waves travelling from the inside of the crystal to the surfaces, assumed at temperature T. The net acoustic energy transfer is $dE_a\approx(U-U_0)v(2A)dt$, where $U(T_p)=[\rho(\nu)\Delta\nu](h\nu/2z)$ is the energy density of the hot lattice oscillators and $U_0(T)$ is a similar quantity for the bath oscillators. We observe the signal decay under the conditions $(z_0-z)\ll z_0$, so that equating dE_z to dE_a yields, approximately,

$$dz/(z-z_0)\approx -2Av\rho\Delta\nu dt/Nz_0^2, \qquad (8)$$

which has the solution for the signal

$$z-z_0=(z_i-z_0)\exp(-t/\tau), \qquad (9)$$

where

$$\frac{1}{\tau}=\frac{2v}{l}\,\frac{12\pi\nu^2\Delta\nu}{v^3c}\coth^2(h\nu/2kT)\text{sec}^{-1}, \qquad (10)$$

where $c=N/Al=$ number of spins per cm^3. If we identify $l/2v$ with T_{ph}, Eq. (10) becomes identical to Eq. (7).

From the linewidths and concentration one obtains values for T_{ph} from the data of Eqs. (6a,b); this is done in SJ, Table III, and the results are shown to be consistent with the interpretation of similar phonon bottlenecks at higher temperatures.

As discussed in SJ, one expects that the observed rate T_b^{-1} will represent T_{1d}^{-1} if $T_{1d}^{-1}\ll\tau_1^{-1}$, i.e., in the absence of a phonon bottleneck; this is the case for Nd in LaMN. On the other hand, if $T_{1d}^{-1}\gg\tau_1^{-1}$ a serious bottleneck exists and one observes $T_b^{-1}=\tau_1^{-1}$, which is evidently the case for Ce in LaMN for the temperatures and concentrations we have used, indicating that the direct process for Ce is considerably stronger than for Nd. From our data, Fig. 4, for the most dilute crystal used, we are able to estimate a lower limit for T_{1d}^{-1} for Ce:

$$T_{1d}^{-1}\gtrsim 20\coth(h\nu/2kT)\text{ sec}^{-1}. \qquad (11)$$

To obtain a theoretical estimate of the direct process for Ce in LaMN, we use the phenomenological approach of Orbach.[9,5] The ground state of trivalent Ce is $4f^1\,{}^2F_{5/2}$, which is split by the crystal field of LaMN into three Kramers doublets (see SJ, Fig. 1): $|a\rangle$ and $|b\rangle$; $|c\rangle$ and $|d\rangle$ at $\Delta_1/k=34$°K; and $|e\rangle$ and $|f\rangle$ at $\Delta_2/k\sim 200$°K. The microwave paramagnetic resonance spectrum[10] of doublet $|a\rangle$ and $|b\rangle$ consists of a single line with $g_{\parallel}=1.8264\pm0.0013$ and $g_{\perp}=0.032\pm0.068$.

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[10] A. H. Cooke, H. J. Duffus, and W. P. Wolf, Phil. Mag. 44, 623 (1953). The g factors we quote are the more recent measurements of H. J. Stapleton at 3 cm and 4.2°K (private communication).

Judd's[11] analysis yields $|A_2^0\langle r^2\rangle|=70$ cm^{-1}, and $|A_4^0\langle r^4\rangle|=30$ cm^{-1} for the crystal field parameters and these wave functions for $z\perp H$ (if one neglects admixtures from the $^2F_{7/2}$ excited level at 2250 cm^{-1}):

$$|a\rangle=0.644[|\tfrac{1}{2}\rangle+|-\tfrac{1}{2}\rangle]+0.288[|\tfrac{5}{2}\rangle-|-\tfrac{5}{2}\rangle], \tag{12a}$$

$$|b\rangle=0.644[-|\tfrac{1}{2}\rangle+|-\tfrac{1}{2}\rangle]+0.288[|\tfrac{5}{2}\rangle+|-\tfrac{5}{2}\rangle], \tag{12b}$$

$$|c\rangle=|-\tfrac{3}{2}\rangle, \tag{12c}$$

$$|d\rangle=|\tfrac{3}{2}\rangle. \tag{12d}$$

The functions $|a\rangle$ and $|b\rangle$ have been chosen to yield $g_{\parallel}=0$; they yield $g_{\perp}=2.14$, which is in only fair agreement with the measured value.

We use SJ [Eq. (18)] to calculate T_{1d}^{-1}; the quantity in the sum we find to be $3.6H^2\times10^3$ cm^{-2} Oe^2, using the procedure outlined in SJ. Taking $v=2.5\times10^5$ cm sec^{-1}, we find $A'\approx5.5$ sec^{-1} deg^{-1}, so that our theoretical estimate for the direct relaxation rate, Eq. (2), becomes

$$T_b^{-1}\approx1.3\coth(h\nu/2kT)\text{sec}^{-1}. \tag{13}$$

This is more than an order of magnitude less than the lower limit set by our data, Eq. (11). On the other hand, we find that $\sum|\langle a|v_n{}^m|c\rangle|^2=2\times10^3$ cm^{-2}, yielding from SJ [Eq. (22a)] a relaxation rate for the Orbach process,[12]

$$T_{1O}^{-1}=3.5\times10^9\exp(-34/T)\text{ sec}^{-1}, \tag{14}$$

which is in rather good agreement with the measured value, Eq. (5). Although one sometimes finds [cf. SJ, Table II] an order of magnitude discrepancy between the measured and theoretically estimated values for other ions, it is usual to find that the calculated *ratios* of two different processes are correctly given. Some possible reasons for the present discrepancy are that the functions, Eq. (12), are not very accurate[13] and that we have neglected the doublet $|e\rangle$ and $|f\rangle$ in calculating T_{1d}^{-1}.

IV. DISCUSSION

These experiments on Ce and Nd in La*MN* crystals bonded with varnish to a cavity cooled down to $T\approx0.25$°K demonstrate that one may perform paramagnetic resonance and even pulsed relaxation time measurements without sample heating problems, other than the phonon bottleneck, at these low temperatures. For Nd, there is not even a phonon bottleneck, as the fit of the data, Fig. 3, shows that all the phonons are maintained at the cavity temperature. For Ce, which has a shorter direct relaxation time, a phonon bottleneck is observed, but this probably could be eliminated by using a Ce concentration of less than 0.01%.

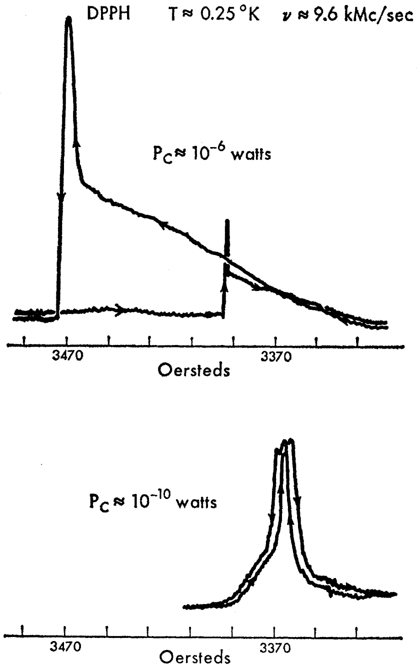

FIG. 5. Paramagnetic resonance absorption of diphenyl picryl hydrazyl at $T\approx0.25$°K at two levels of cavity power P_c. The arrows indicate the direction of dc magnetic field sweep; the hysteresis effect is due to the temperature dependence of the local field in the sample.

On the other hand, for magnetically concentrated samples the heating problem may not be negligible. For example, a speck (~1 mg) of diphenyl picryl hydrazyl (DPPH), stuck to the cavity with a spot of silicone vacuum grease, showed the resonance behavior of Fig. 5 at $T\approx0.25$°K. At a cavity power $P_c\approx10^{-10}$ W the paramagnetic resonance absorption (observed without field modulation directly from the dc output of the superheterodyne receiver) displays a slight shift about $H_0=3770$ Oe, depending on the direction of field sweep. At a cavity power $P_c\approx10^{-6}$ W this effect is very pronounced: as the resonance is swept through with H increasing, the absorption does not appear at H_0 but is pushed up about 100 Oe and then suddenly snaps out; with H decreasing the absorption appears suddenly at about 20 Oe above H_0. Although this hysteresis behavior may be attributed to ferro- or antimagnetism, it also has a simpler classical explanation. If we assume a paramagnetic sample obeying Curie's law, then the applied field for resonance will be

$$H_0=\frac{h\nu}{g\beta}\left[1-\left(\frac{4\pi}{3}-N\right)\frac{C}{T}\right], \tag{15}$$

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[11] B. R. Judd, Proc. Roy. Soc. (London) **A232**, 458 (1955).

[12] In order to be consistent with our calculation of the direct process we have used the procedure of SJ to calculate the Orbach process, obtaining a value in reasonable agreement with the original calculations of Finn, *et al.*, reference 5: $T_{1O}^{-1}\approx10^{10}\times\exp(-34/T)$ sec^{-1}. The main differences are that we use SJ, Eq. (15) to estimate the crystal field parameters; and we take *all* nonvanishing matrix elements, added incoherently as in SJ, Eq. (16).

[13] *Note added in proof.* R. Orbach (private communication) has obtained improved wave functions and crystal field parameters which cast doubt on ones used in our calculation. Unfortunately, the improved functions do not yield a value of T_1 in better agreement with the experimental result.

where C is Curie's constant and N is the demagnetization factor, determined by the sample shape. Thus as the temperature is lowered to $T=0.25°\text{K}$ the resonance field H_0 may be appreciably shifted to lower fields. If microwave power is applied and if the sample is in poor thermal contact with the cavity, the resonant absorption will heat the sample and move the line back toward higher fields, so that an increasing field sweep will appear to push the line up to a limiting field, where the line then snaps out and returns to H_0. This type of behavior is to be generally expected in microwave paramagnetic resonance absorption below 1°K under conditions of sample heating.

We have also considered the possibility that this shift is due to the Overhauser effect,[14] but it is an order of magnitude too large, the hyperfine coupling constants being only a few Mc/sec for the protons[15] and the N^{14} nuclei[16] in DPPH.

Returning to Nd in La*MN*, we note that the relaxation time T_{1d} is rather long at $T\approx 0.25°\text{K}$; from the expected field dependence $T_{1d}^{-1}\propto H^5\coth(g\beta H/2kT)$ of Eq. (2) we estimate $T_{1d}\approx 130$ sec at $H\approx 10^3$ Oe, and $T_{1d}\approx 10^6$ sec at $H\approx 10^2$ Oe. This is so long that the Nd spins are effectively isolated from the bath; thus, low spin temperatures could be obtained without a heat switch by, say, starting with a dilute Nd, La*MN* crystal bonded to a reservoir at $T_i=0.1°\text{K}$ and in $H_i=10^4$ Oe. The measured linewidth of the Nd resonance is 2.5 Oe in a 1% crystal, so that a final spin temperature $T_f\approx 2.5\times 10^{-5}°\text{K}$ may be achieved and maintained if the field H_i were reduced to zero with an appreciable reduction occurring in the first few seconds. Spin temperature mixing experiments between electron spins and other electron spins, and also between electron spins and nuclear spins, would be feasible, in analogy to the experiments[17] with nuclear spins.

We wish to acknowledge with thanks the assistance of N. Ford in the performance of the experiments and the design of the microwave switch; the helpful comments from N. Kurti and L. D. Roberts on the cryostat design; and the informative discussions with B. Bleaney, N. Bloembergen, P. L. Scott, and R. Orbach.

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