PROGRESS REPORT

EXPERIMENTAL AND THEORETICAL STUDIES
OF MAGNETIC RESONANCE AND RELAXATION
for period September 1, 1967 to August 15, 1968

UNIVERSITY OF KANSAS
ATOMIC ENERGY COMMISSION
Contract No. AT(11-1)-1488

Technical Report COO-1488-13

LEGAL NOTICE
This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
Progress is reported on experimental projects in nuclear and electronic spin wave excitation and improvement of fast pulse techniques. Theoretical work on nuclear spin wave relaxation, relation between spin-lattice relaxation and line width in the Orbach process, and calculation of magnetic resonance linewidth is reported. Period covered is from September 1, 1967, to August 15, 1968. Variation of the product of electronic and nuclear spin wave relaxation rates in RbMnF$_3$ has been measured as a function of wave vector and temperature between 2.4°K and 10.5°K. A minimum is found at a given wave vector which increases linearly with temperature. A broad band helix is being incorporated into the spectrometer for future high-power fast-pulse work employing a 1 kW travelling wave tube currently on order. Theoretical treatment of nuclear spin wave relaxation rates shows them to be considerably less than expected on the basis of second moment of the Suhl-Nakamura interaction. In this way we explain the anomalously narrow NMR line width in RbMnF$_3$ and long lifetimes of nuclear spin waves as inferred from our parallel pumping data. Calculation of moments of the resonance line by random phase approximation (Tyablikov decoupling) appears to be totally inadequate for explaining line width in nickel near the Curie point. The line width of isolated paramagnetic ions has been derived for spin lattice interaction via the Orbach process. Observed discrepancy between line width and spin lattice relaxation rate can be accounted for by this calculation only if there is phonon bottlenecking, which we suggest is the case. Four technical reports have been issued during the period of this report. Three have been accepted for publication in Physical Review. The fourth is a thesis not intended for publication. Three other journal articles have appeared in print during this period. These were submitted during the previous contract period.
I. Excitation of Nuclear and Electronic Spin Waves by Parallel Pumping.

We have made extensive measurements of simultaneous excitation of nuclear and electronic spin waves in Rb\textsubscript{2}InF\textsubscript{3} between 2.4°K and 10.5°K. This is a continuation of the preliminary work reported in the Progress Report of August 31, 1967, and published in J. Appl. Phys. 39, 324 (1968) [Proceedings of 1967 International Congress on Magnetism]. The results, which will be submitted for publication in the near future, give products of nuclear and electronic spin wave relaxation rates as a function of wave vector for the temperature range studied and are discussed below.

First consider the possibility of detecting electronic spin waves alone. In the Proposal for the current contract period it was suggested that in the vicinity of 10°K it might be possible to do parallel pumping in which a pair of electronic spin waves is excited. This would be useful since then the electronic relaxation rate $n_e$ could be measured. In the simultaneous excitation of electronic and nuclear spin waves only the product $n_e n_n$ of electronic and nuclear relaxation rates can be measured from observation of threshold fields. Temperatures higher than that of liquid Helium are required because the electronic frequency $\omega_e(k)$ must be sufficiently low to satisfy

$$\omega_e(k) = \omega/2$$

for excitation of electronic spin waves alone by a driving field of frequency $\omega$. The minimum of $\omega_e(k)$ occurs at wave vector $k = 0$ for a given dc field $H$, and the smallest value of $\omega_e(0)$ occurs when $H$ is at the minimum value ("flopping" field) required to bring the sublattice magnetizations into the plane perpendicular to it. At this value of $H$ we have

$$\omega_e(0) = \sqrt{2\gamma H_H E_H}$$
where $H_E$ and $H_H$ are the exchange and hyperfine fields, respectively, seen by the electronic spins and $\gamma$ is the gyromagnetic ratio. In particular, $H_H$ varies as $1/T$ since it is proportional to the nuclear polarization. For a pumping frequency $\omega/2\pi = 8.5$ GHz as used in our experiments, the temperature must be at least 7°K in order to satisfy (1).

At 10°K there should be a range of dc fields for which (1) can be satisfied. However, the effect can be observed readily only if it has a threshold lower than that required for the simultaneous excitation of electronic and nuclear spin waves, for which the frequencies satisfy

$$\omega_e(k) + \omega_n(k) = \omega$$

(2)

The reason may be seen from Fig. 1 which is a sketch of critical r-f field $h_c$ vs. $H$. Referring to Fig. 1, the field $H_c$ is the flopping field, $H_1$ is the maximum field for which it is possible to satisfy (1), and $H_2$ is the maximum field for which it is possible to satisfy (2). The dashed curve represents what one might expect to see if the process (1) had the lower $h_c$. In this case $h_c$ should show a sharp rise near $H_1$ since for $H > H_1$ the process (1) would be forbidden. If, on the other hand, process (2) had the lower threshold, then the solid curve would be appropriate and there would be no striking behavior near $H_1$. This is because $h_c$ measures the first point at which strong absorption and pulse distortion are seen. Either of the two parallel pumping processes will produce these effects.

Our experimental results are consistent with the solid curve of Fig. 1. The difference between $H_1$ and $H_c$ is about 150 Oe at 10.5°K for the frequency and crystal orientation ($H$ in [100] direction) we use. This should be a large enough region for effects associated with the dashed curve to show up without any trouble. We conclude, therefore, that process
Figure 1. Critical rf field $h_c$ vs. dc field $H$.

Solid line: behavior if simultaneous excitation of nuclear and electronic spin waves has lower threshold.

Dashed line: behavior if excitation of two electronic spin waves has lower threshold.
(2), the simultaneous excitation of nuclear and electronic spin waves, actually has a lower threshold than the more familiar parallel pumping of two electronic spin waves (1). This is a consequence of the extremely low relaxation rates of nuclear spin waves, as discussed in Sect. II.

We have derived an expression for $h_c$ in process (1) for a flopped antiferromagnet. The result is

$$h_c = \frac{2n_e \omega}{\gamma^2 H} \quad (3)$$

From the fact that at 10.5°K in the field $H_1 = 2600$ Oe (See Fig. 1) we measure a critical field of 2.45 Oe for process (2), we conclude from (3) that

$$n_e \approx 1.9 \times 10^7 \text{ sec}^{-1}$$

since if $n_e$ were much smaller than this value, behavior such as the dashed curve on Fig. 1 should be expected. This number for $n_e$, which corresponds to a half-width of 1.06 Oe, would be for $k \to 0$ and an electronic frequency of 4.25 GHz at 10.5°K. Cole and Courtney inferred $n_e = 6.7 \times 10^7 \text{ sec}^{-1}$ from saturation measurements of AFMR. Our lower limit is thus well below their value, but their data are for $\omega_e/2\pi = 8.075$ GHz and $T = 4.2$°K, so some caution must be used in attempting a comparison.

Fig. 2 shows a typical curve of $n_e n_n$ vs. $k$ at a given temperature. This curve is obtained from an experimental curve of $h_c$ vs. $H$ as in Fig. 1 by 1) determining $k$ from the relation $\omega_e(k) + \omega_n(k) = \omega$ where $\omega_e(k)$ and $\omega_n(k)$ are given in terms of $H_E$, $H_N$ and $H_A$ (anisotropy field), all of which are known for RbMnF$_3$, and the applied field $H$ and 2) determining $n_e n_n$ from the relation we have derived$^2$ for $h_c$ in process (2) in terms of $H_E$, $H_N$, $H_A$.
Figure 2. Relaxation rates vs. wave vector $k$ at 6.4°K, $\omega/2\pi = 8.5$ GHz.

- $\eta_e \eta_n$, experimental
- $\eta_n$, theoretical
- $\eta_e$, inferred from $\square$ and $\triangle$
Figure 3. Wave vector at which product of relaxation rates is a minimum vs. temperature.
Figure 4. Product of relaxation rates at minimum value vs. temperature.
H and \( \omega \) for a flopped antiferromagnet. The feature of a minimum in \( n_e^* n_n \) vs. \( k \) shows up clearly at every temperature except the highest, 10.5°K. At 10.5°K there is some suggestion of a minimum, but the \( k \) for which it occurs is at the end of our range. At temperatures above 10.5°K we are unable to see parallel-pumping instabilities, presumably because the critical r-f fields are beyond the reach of our present apparatus.

In Fig. 2 we also show curves for \( \eta_n \) and \( \eta_e \) separately. These are obtained by taking the theoretical value we have obtained for \( \eta_n \) (Sect. II) and dividing it into the experimental product \( n_e^* n_n \) to get \( n_e \).

Figs. 3 and 4 show, respectively, as functions of temperature the value of \( k \) at which the minimum occurs and the value of \( n_e n_n \) at that minimum. The \( k \) corresponding to the minimum is seen to increase linearly with temperature while \( n_e n_n \) at the minimum decreases very slightly with temperature until 9°K and then starts to rise more rapidly. Roughly we can describe these effects by writing

\[
\eta_e n_n = \frac{\alpha K}{T} + \frac{\beta T}{k}
\]

where \( \alpha \) and \( \beta \) are appropriate constants. Eq. (4) then predicts \( n_e n_n \) to be a minimum for \( k = \sqrt{\beta/\alpha} T \) at which point \( n_e n_n = 2 \sqrt{\alpha/\beta} \), independent of temperature.

Our theory of nuclear spin wave relaxation (Sect. II) gives no dependence such as (4) for \( \eta_n \). A corresponding theory of electronic spin wave relaxation in a flopped antiferromagnet does not yet exist. Dr. R. M. White of Stanford University, with whom we have been in close contact regarding these results, is currently investigating this latter theoretical problem. Sect. II contains some further remarks on the comparison between theory and experiment.
Immediate plans call for further analysis of the data of Figs. 2-4 so that we may at least present an accurate phenomenological description of relaxation rates vs. temperature and wave vector. There also may be some hope that we can measure $n_e$ and $n_n$ separately by fast pulse techniques with our new 1 kw travelling wave tube scheduled for arrival in September, 1968. (See Sect. III).

II. Theory of Nuclear Spin Wave Relaxation

We have developed a theory of relaxation of nuclear spin waves which appears to give reasonable agreement with experiment. It is Technical Report COO-1488-10 and has been accepted for publication in Physical Review (scheduled for Sept. 10, 1968, issue). Briefly, the idea is that if there is a well-defined nuclear spin wave spectrum so that the spread of frequencies between $k = 0$ and the maximum $k$ in the first Brillouin zone is large compared with the root mean square frequency width of a given spin wave, then only a small portion of the total number of spin waves can interact in energy conserving processes. As a result, relaxation rates can be calculated by means of transition probabilities and are very much less than would be inferred from the second moment $<(\omega - \omega_k)^2>$. Details may be found in COO-1483-10 and thus are not repeated here.

An immediate consequence of the theory is that it explains why the observed NMR line width in RbMnF$_3$ is an order of magnitude less than calculated on the basis of the second moment of the Suhl-Nakamura interaction. We calculate a relaxation rate for the $k = 0$ nuclear spin wave of about $10^3$ sec$^{-1}$ at 4.2°K. This is two orders of magnitude less than the NMR width, which is not particularly disturbing since the NMR width
is undoubtedly influenced by inhomogeneities. It is well established\(^4\) that the AFMR width in Rb\(\text{MnF}_3\) is due almost entirely to random strains.

If our value \(10^3 \text{ sec}^{-1}\) for \(\eta_e\) is combined with Cole and Courtney's figure \(6.7 \times 10^7 \text{ sec}^{-1}\), we come very close to the experimentally observed product \(\eta_n\eta_e\) at \(k = 0\). However, agreement is worse at higher values of \(k\) since, at \(4.2^\circ\text{K}\), theory gives \(\eta_n = 2.6 \times 10^4 \text{ sec}^{-1}\) for \(k = 10^5 \text{ cm}^{-1}\). This would then require \(\eta_e \sim 3.5 \times 10^6 \text{ sec}^{-1}\) to fit our experimental results for \(\eta_e\eta_n\).

Before absolute judgment can be rendered on quantitative validity of our theory, it is necessary to know \(\eta_n\) by itself either through determination of \(\eta_e\) as well as \(\eta_e\eta_n\) or by parallel pumping of nuclear spin waves alone. Cole and Courtney's inferred value for \(\eta_e\) may be subject to some error since several assumptions regarding distribution of inhomogeneities and dependence of \(\eta_e\) on \(k\) had to be made by them in order to arrive at a figure for \(\eta_e\) from the observed decline of AFMR susceptibility at high power levels.

Also it is likely that a proper theory of relaxation rates in Rb\(\text{MnF}_3\) must include inhomogeneities even for parallel pumping measurements. In the meantime, however, our theory seems to be at least qualitatively correct and explains why NMR line width is so much narrower than predicted by second moment methods. Previously, the narrowness of the NMR line in Rb\(\text{MnF}_3\) had been regarded as a major anomaly. We also are able to see why nuclear spin wave relaxation rates can be considerably less than had originally been predicted so that excitation of nuclear spin waves by parallel pumping requires nowhere near as great an r-f field as had been supposed.\(^5\)

III. High Power Fast Pulse Spectrometer

As mentioned in the Proposal for the current contract period and in the Progress Report of August 31, 1967, our spectrometer was incapable
of producing pulses of sufficiently high power in low Q circuits in order to perform several of the desired experiments. Such experiments include observation of exchange-lattice relaxation in concentrated paramagnetic salts and measurements of spin wave relaxation rates from transient decay of parallel-pumping signals. They require high power in order to saturate the system or produce the necessary nonlinear coupling, but at the same time high Q resonant cavities cannot be employed since their response time is sufficiently long to obscure the transients under investigation.

Some steps have been taken to improve the situation. First, a 1 kw travelling wave tube and supply has been ordered from Litton Industries. Delivery is scheduled for September, 1968. Since our present tube puts out 10 watts, this means that r-f h fields can be increased by a factor of 10 so that our capability for observing nonlinear and saturation effects will be greatly enhanced. It might then, among other things, be possible to investigate nuclear spinwaves at temperatures above 10.5°K. In terms of Q, the additional factor of 100 in power means, for example, that the same h will be obtainable in a cavity whose Q is 50 as can now be obtained only with a Q as high as 5000.

Secondly, we have incorporated a helix into the spectrometer. A helix has the advantage of being very broad band so that its response time presents no limitation. At the same time filling factors close to unity can be attained so that strong resonance signals are still possible. In this respect the helix is preferable to a very low Q cavity since in the latter case signal strength can be a definite problem. The particular helix we are using is 0.07 inches in diameter wound with 20 turns of No. 22 copper wire. The final two turns are soldered together to form a short. It is placed outside the waveguide and coupling accomplished by a probe. Tests have been run on parallel pumping in a 0.05 inch diameter sphere
of yttrium iron garnet. Field strength in the helix is calibrated by comparing the power level at which threshold occurs when the sample is in the helix with the power required when the sample is placed in a cavity of known dimensions and $Q$. We find, for a given power level, the helix to give about the same field as the maximum obtainable in a critically coupled $TE_{101}$ cavity made from X-band waveguide with a loaded $Q$ of 5000.

In order to utilize the broadband character of the helix we have found it essential to match all reflections. We use a bridge detection scheme, and the problem of pulse reflections is illustrated in Fig. 5. If the detected signal is to be zero when the bridge is balanced, the pulses reflected from the helix and variable short must return to the magic-tee junction at the same time. If they do not, then a large unbalance occurs at the beginning and end of the pulse even though exact balance can be achieved in the center of the pulse, as shown in (a). Only if the lengths $L_1$ and $L_2$ are equal can the bridge be balanced at the ends of the pulse. Also any small intermediate reflections in the length of waveguide $L_1$ will have to be compensated for by corresponding reflections in $L_2$.

The above considerations are no problem in cw measurements or even in studies of transients of the order of $10^{-6}$ seconds and longer. But we are interested in relaxation times which may be as short as 20 nanoseconds, and delays of close to this length are encountered in the wave guide system since the length $L_2$ is about 8 feet. To compensate for this, we have placed extra waveguide in the system to make $L_1 = L_2$. Also slide screw tuners are placed in both arms to balance out reflections. The process of balancing out reflections has proved to be quite tedious, but it is nearly completed, and it appears that satisfactory results can be obtained.
If there is sizeable unbalance as in (a) of Fig. 5, then transients associated with the resonance signal will be obscured.

The balanced helix system will first be used in an attempt to observe transients associated with simultaneous excitation of nuclear and electronic spin waves in RbMnF$_3$. He also hopes to investigate exchange-lattice relaxation. No efforts have been made in this regard during the present contract period since we prefer to wait for the 1 kw travelling tube, with which chances for success should be greatly increased.

IV. Theory of Temperature Dependence of Ferromagnetic Resonance Linewidth

We have completed calculations of the second and fourth moments of the magnetic resonance line in a face centered cubic lattice of localized spin 1/2 moments. Comparison has been made with experimental results on line width in nickel$^7$ and agreement is extremely poor.

According to the method of moments$^8,^9$, exchange-narrowed line width $\Delta H$ is given by

$$\Delta H \propto (M_2)^{3/2}(M_4)^{-1/2}$$  \hspace{1cm} (5)

where $M_2$ and $M_4$ are the second and fourth moments, respectively, of the resonance line. Eq. (5) may also be written

$$\frac{(M_4)^{-1/2}(M_2)^{3/2}}{<S_x^2>} = \frac{<g^+g>^{3/2}}{<g^+g^{1/2}>}$$  \hspace{1cm} (6)

where

$$g = \frac{1}{\hbar} [H', S_x]$$  \hspace{1cm} (7)

in which $H'$ is the line-width-inducing perturbation taken to be pseudo-dipolar coupling for Ni, and $S_x$ is the total x-component of spin in the
Figure 5. Schematic of incident and reflected pulses in microwave circuit employing helix. Pulse incident from above splits in two at magic tee junction. One pulse travels distance $L_1$ to variable short and is reflected back to junction. Other travels distance $L_2$ to helix and is reflected back to junction. Detector measures difference between reflected pulses at junction.
crystal. Time dependence of \( g \) is governed by the exchange interaction \( H_{\text{ex}} \) so that

\[
g = \frac{i}{\hbar} [H_{\text{ex}}, g]
\]  

(3)

As discussed in the Proposal for the current contract period, and in the Progress Report of August, 1967, our procedure is to compute \( \langle g^+g \rangle \) and \( \langle g^+g \rangle \), which involve, respectively, four spin and six spin correlation functions by an extension of the high-density approximation of Stinchcombe et al.\(^\text{10}\) In this method four and six spin averages are decoupled into products of pair correlation functions, and the pair correlations are evaluated by random phase approximation (Tyablikov decoupling).

We find that \( \langle g^+g \rangle \) shows a marked increase as \( T \) approaches \( T_c \) either from below or above. The quantity \( \langle g^+g \rangle \) goes negative at \( T_c \), which of course is absurd. One's first reaction to this latter result would be to assume numerical error in the calculation. We are confident, however, that no error exists and that this is a real effect of the random phase approximation. During the course of these calculations an important paper was published by Liu and Siano\(^\text{11}\) which showed that, for spin 1/2, the random phase approximation yields a negative value for the specific heat at \( T_c \). The difficulty seems to be that the random phase approximation gives a reasonable description of long range correlations but fails for the short range correlations. (It is the short range order which determines internal energy and hence specific heat). Investigation shows that \( \langle g^+g \rangle \) does involve short range correlations only. At first glance it appears that \( \langle g^+g \rangle \) contains long range correlations, but the dipole factors may limit the importance of these terms. If this is so, then \( \langle g^+g \rangle \) may similarly be in gross error near \( T_c \). We are studying this
point at the moment.

Since random phase approximation obviously gives an incorrect calculation of \( \langle \mathbf{g}^+ \mathbf{g} \rangle \), we have attempted to compare theory with experiment by assuming \( \langle \mathbf{g}^+ \mathbf{g} \rangle \) is temperature independent near \( T_C \). This is reasonable since \( \langle \mathbf{g}^+ \mathbf{g} \rangle \) involves short-range correlations only. We then find the temperature dependence of \( \Delta H \) to be proportional to \( \langle \mathbf{g}^+ \mathbf{g} \rangle^{3/2} / \langle S^2 \rangle \). This quantity is plotted vs. \( T/T_C \) in Fig. 5 along with Salamon's results\(^7\) for Ni. Theoretical curves are for dc fields of 6 kOe, which is approximately the field used by Salamon, and 80 kOe for comparison. Experimental and theoretical curves have been normalized to agree at \( 0.99 T_C \).

The experimental linewidth decreases by about a factor of 14 as \( T \) is lowered from 1.2 \( T_C \) to \( T_C \), but the theoretical linewidth changes much less, and the most pronounced effect, at 6 kOe, is an increase at \( T_C \). This increase is washed out at 80 kOe, as can be seen.

Clearly there is not satisfactory agreement. It is interesting to compare Salamon's results with those of Ford and Jeffries\(^12\) on \( K_2CuCl_4 \cdot 2H_2O \). Ford and Jeffries find linewidth to decrease by only about a factor of 1.5 between 1.2 \( T_C \) and \( T_C \) (\( T_C = 1.10^\circ K \)) so that their data would appear as nearly constant on the scale of Fig. 6. Magnetic field energy is comparable to \( k_B T_C \) (\( k_B = Boltzmann's \) constant) in their experiment while it is much less than \( k_B T_C \) in Salamon's. In fact a field of about 1800 kOe in Ni would be required to give the same ratio of \( \mu H/k_B T_C \) (\( \mu = \) magnetic moment) as used in \( K_2CuCl_4 \cdot 2H_2O \). There may be some chance that random phase approximation will give reasonable results in high fields and can be used to interpret the measurements of Ford and Jeffries. This point and other possible methods of approaching the problem in Ni are discussed in the Proposal.
Figure 6. Line width in Ni vs. temperature near Curie temperature $T_c$.

$\Theta$ - experiment ($H \approx 7$ kOe)

solid lines - random phase approximation calculation of

$\langle g^+ g \rangle^{3/2} / \langle S_x \rangle^2$ for two different dc fields.
V. Time Correlation Functions of Spin Operators in Finite Heisenberg Linear Chains.

Dr. Fernando Carboni's work (Technical Report COO-1488-4) on time correlation functions in finite linear chains has been extended somewhat during the present contract period. Results have now been obtained for open chains (lack of periodic boundary conditions) containing as many as nine spins. The matrices which have to be diagonalized in this case are considerably larger since translational invariance cannot be used to introduce a new set of quantum numbers. There are no basic differences between results in open and closed chains of 9 spins. This is gratifying since it reliably be extrapolated to the infinite chain limit. In fact it appears that convergence to the $N \to \infty$ limit is more rapid in open chains than in closed chains.

These new calculations have been included with the ones reported earlier in a complete paper which has been accepted for publication in Physical Review (Technical Report COO-1488-11). In addition to presenting the results, we have made detailed comparison with various approximate theories of time correlation functions and with numerical calculations performed by Windsor on classical spin systems. Agreement with the classical results is extremely good. This is encouraging since classical calculations can be (and have been) performed on three-dimensional systems as well, whereas our quantum mechanical methods cannot be extended beyond one dimension at least with present day computers. There seems to be no reason why classical and quantum calculations should not agree in three dimensions given that they do in one dimension.
Since time correlation functions are needed for calculating neutron scattering cross sections as well as for magnetic resonance linewidths, results obtained by us can have a broad range of applicability. Agreement with experiment has already been shown to be excellent for magnetic resonance line width. It is hoped that neutron scattering data on linear chain salts may come available to provide a further test.

VI. Spin Lattice Relaxation and Line Width due to Orbach Process.

In a joint effort with Professor J. W. Culvahouse of this department we have considered relaxation of paramagnetic ions by the Orbach relaxation process. This is the material contained in Technical Report COO-1488-12 which has been accepted for publication in Physical Review. Our interest stemmed from experiments of Stapleton and Brower on small concentrations of Ce\textsuperscript{3+} in Lanthanum Magnesium Nitrate. They found both the line width $\Delta H = 1/\gamma T_2$ and spin-lattice relaxation rate $1/T_1$ have the characteristic temperature dependence of the Orbach process, but that $T_1$ is an order of magnitude greater than $T_2$. The question arose whether it was possible to have this big a difference between $T_1$ and $T_2$ for a case in which all the broadening is due to spin-lattice coupling via the Orbach process.

We reformulated the Orbach theory using a density matrix approach whereby $T_2$ could be calculated as readily as $T_1$. Previous studies of the Orbach process considered $T_1$ only. Results showed that in general $T > T_2$ but that $T_1 = T_2$ provided $B_1 = B_2$ where, for a pair of Kramers doublets $|\pm p/2\rangle$ and $|\pm q/2\rangle$, $B_1$ is the probability for the transition $|p/2\rangle \rightarrow |q/2\rangle$ and $B_2$ is the probability for the transition $|p/2\rangle \rightarrow |-q/2\rangle$. Symmetry arguments for a Kramers ion in a site of trigonal symmetry were then used to show that in fact $B_1 = B_2$ for the dc field applied perpendicular
to the axis of symmetry, as was the situation in Stapleton and Brower's work.

It thus appeared that the result $T_1 \gg T_2$ was incompatible with the simple Orbach process. We have proposed that the explanation is bottlenecking of a certain class of phonons even at low concentrations. Bottlenecking lengthens $T_1$ but, as we show in the paper, has no effect on $T_2$. Hence it is then possible to have $T_1 \gg T_2$ even with $B_1 = B_2$. Our study lends further support to a wide variety of evidence which has recently been put forward in favor of a near-universal bottlenecking in the Orbach process.

VII. Study of Spin Wave Properties from Harmonic Generation in YIG.

Dr. John Bierlein became the second person to receive a Ph. D. under this contract in December, 1967. His thesis is contained in Technical Report COO-1488-9. The work is described fully in that Report and also in the Progress Report of August 31, 1967. A preliminary account was published some time ago, and we hope to submit a more complete paper in the near future.

VIII. Publications and Reports

The following is a list of Technical Reports which have been issued during the period of the present contract and their publication status.


COO-1488-11: "Time Dependence of Spin Operators in Finite Heisenberg Linear Chains" by Fernando Carboni and Peter M. Richards, (Physical Review, accepted for publication)

COO-1488-12: "T₁ and T₂ for Orbach Relaxation Processes" by J. W. Culvahouse and Peter M. Richards (Physical Review, accepted for publication).

The following papers have been published since the last Progress Report. They were all prepared during the previous contract period.


References


15. H. Stapleton and K. Brower, to be published.