MASTER

RECEIVED BY DTIC AUG 21 1970

FINAL REPORT

AEC CONTRACT AT(11-1)1488

[ON DYNAMIC ASPECTS OF CONCENTRATED MAGNETIC SYSTEMS]

Peter M. Richards
Department of Physics
University of Kansas
Lawrence, Kansas

TECHNICAL REPORT C00-1488-21

June, 1970

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or 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.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

fy

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.

During the five years (June 1, 1965, to May 31, 1970) of this contract, significant results, both experimental and theoretical, have been obtained on dynamic aspects of concentrated magnetic systems. On the experimental side, the prime success has been in excitation of spin waves and observation of their lifetimes through novel fast-pulse techniques. Most significant has been the Ph.D. thesis work of L. W. Hinderks in which nuclear spin waves were excited and measured for the first time in any laboratory. Theoretical effort has mostly been concentrated on time dependence of exchange-interacting spins and how this relates to observed magnetic resonance linewidths. If one is to single out a most important work in this area, it would probably be the Ph.D. thesis of F. Carboni in which time dependence was computed exactly for finite linear chains of spin $1/2$ particles. This research represented the first such exact quantum mechanical calculation on a system sufficiently complicated to show the expected characteristics of a real magnetic substance. It gave good agreement with experiment whereas agreement had been very poor previously and has stimulated considerable effort by researchers at other institutions.

The specific works accomplished are discussed below with references made to the appropriate Technical Reports and publications. Since practically all the research does appear in these reports and publications, the discussion is brief and emphasizes the significance rather than results or methods. The work which does not yet appear in report form is that of students whose Ph.D. research was begun under this contract and has not progressed sufficiently to warrant a full report at this time.

A. Experiment

1. Harmonic Generation and Spin Wave Excitation in Yttrium Iron Garnet.

(Reports 3, 9, 18; Publications 1, 11)

This work, which represented the Ph.D. thesis of J. D. Bierlein, had two prominent features. First, it showed that harmonic generation could be useful as a tool in studying the distribution of excited spin waves, particularly their phases. Secondly, it gave conclusive evidence that definite phase relations exist among the excited spin waves which must be properly taken into account in order to explain the data. This is quite significant since previous workers had generally assumed that random phases and "Golden-Rule" type arguments could account for spin waves not directly excited by the external r-f fields. Bierlein's research showed this definitely not to be the case. Strong coherence effects were demonstrated to be present, giving further evidence that phases can be important in quantum systems.

2. Nuclear Spin Waves (Reports 7, 14, 17, 20; Publications 4, 8, 10)

The first-ever excitation of nuclear spin waves with non-zero wave vector was accomplished under the Contract. Strong evidence was given for the existence of nuclear spin waves as well-defined collective excitations. Elementary excitations form the basis of our understanding of solid state physics, and any discovery such as this which adds to the list of "fundamental particles" of solid state physics (such as phonons, magnons, polarons, etc.) is obviously important. Nuclear spin waves are a particularly interesting form of excitation which should provide a rich basis for future study. This is because

they represent a convenient means of studying collective motion in an essentially disordered system. The nuclear moments have only 1% or less relative polarization, but in spite of this large disorder among the nuclei, well-defined spin-waves are able to propagate with long lifetimes. Nuclear spin waves may possibly prove to be useful for devices operating in the UHF range. If so, our work which first detected nuclear spin waves and measured their lifetimes will have to be considered one of the foundations.

3. Measurement of Spin Wave Lifetimes from Transient Decays.

One of the original goals of the research under this contract was to measure very short spin-lattice relaxation times of the order of 10^{-8} seconds, in paramagnetic resonance. This was abandoned as unfeasible with available equipment and techniques. However, the general interest in short response times has persisted, and both Bierlein and Hinderks were able to make direct transient decay measurements of spin wave lifetimes. Bierlein resolved decay times as short as 2×10^{-8} sec. Hinderks worked on a much longer time scale with nuclear spin waves but was able to establish an important fact: Lifetimes of spin waves excited by parallel pumping can be measured by observing the transient decay of the parallel-pump signal and correlated with critical r-f fields. This is significant because previous measurements have made use of the relation

$$(\eta_1 \eta_2)^{1/2} = K h_c \quad (1)$$

where η_1 and η_2 are relaxation rates associated with the two spin waves excited in the parallel pump experiment, h_c is the critical r-f field required to produce

the excitation, and K is a constant dependent on parameters of the spin wave spectrum. Relaxation rates were then determined from the measured h_c and the assumedly known K . For excitation of two nuclear or two electronic spin waves, $\eta_1 = \eta_2$; but for simultaneous electronic-nuclear excitations, $\eta_1 \neq \eta_2$, and (1) is not sufficient to determine individual relaxation rates.

Transient measurement is desirable because (a) it gives a direct determination of η as opposed to the more indirect Eq. (1), (b) it can serve as a check on the validity of (1) and the assumed value for K and, (c) for the simultaneous electronic-nuclear excitation it can provide a means of sorting out η_1 and η_2 . This is because the transient decay rate is $\eta_1 + \eta_2$; so a measurement of $\eta_1 + \eta_2$ combined with $\eta_1\eta_2$ from (1) can give η_1 and η_2 separately.

Another graduate student, B. Adams, has pursued the work of Bierlein and Hinderks and is measuring transient decay of parallel-pumped spin waves in YIG. This is a first step in a broad program of measuring the short lifetimes of electronic spin waves. He has solved many of the difficulties encountered in our earlier attempts at spin-lattice relaxation. Sophisticated use of signal averaging has eliminated the undesirable "spikes" associated with the ringing of the microwave circuitry, and use of non-resonant helices has provided a broadband system which at the same time is fairly sensitive. Two major items of equipment purchased on the contract, a 1 kW high power travelling wave tube (TWT) and a low-noise TWT for detection, are crucial to this project.

B. Theory

1. Dynamics of Heisenberg Linear Chains (Reports 4, 5, 6, 11; Publications 2, 3, 6).

Time correlation functions of spin operators interacting via a Heisenberg exchange interaction form the input for any calculation of inelastic neutron scattering (in magnetic materials) or magnetic resonance linewidths and are fundamental to a basic understanding of ordered magnetism. Prior to the Ph.D. thesis of F. Carboni there were several approximate theories but nothing of an exact nature. Carboni took a model which was soluble - numerically - but still contained a sufficient number of spins that the expected random and diffusive natures of the correlation functions showed up in the results. His calculation was for a one-dimensional system; so it is not immediately obvious how relevant it is to three-dimensional nature. However, two strong points can be made in this regard. First, there do exist substances which are one-dimensional as far as magnetic properties are concerned. Thus, it was possible to make direct comparison of his calculations with experiments on magnetic resonance linewidth. Agreement proved to be very good and removed what had appeared to be a major discrepancy in correlating linewidth and specific heat measurement in the linear chain salt $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ (copper tetramine sulfate monohydrate, referred to as CTS).

Secondly, one of the main virtues of having an exact numerical result is that it can be used to check the validity of approximate analytic formulae. Although the exact result can be obtained only in one dimension with present-day

computers, the approximations are by no means restricted to one dimension. In fact, most of the approximations are expected to be poorest in one-dimension. Thus, if an approximate theory works in one dimension, it might be expected to work in three dimensions as well. Carboni's results, therefore, have provided a standard against which approximations can be tested. Such tests have been made, and it appears¹ that one can now say that of the many possible techniques, the method of generalized diffusivity gives perhaps the best handle on correlation functions. It has been possible to make this statement only because of the existence of Carboni's exact work.

The divergent properties of the linear chain correlation functions at zero frequency require a reformulation of the theory of magnetic resonance linewidth in substances such as CTS. This has been done by C. McElwee as part of his Ph.D. thesis work which will be completed in September, 1970. A preliminary report of this work is enclosed as Technical Report C00-1488-19. His results seem to show that interchain coupling which removes the one-dimensionality may be an order of magnitude larger than previously estimated.

¹R. A. Tahir-Kheli and D. G. McFadden, Phys. Rev. 178, 800 (1969).

2. Temperature Dependence of Ferromagnetic Resonance Linewidth (Reports 2, 16).

There has been considerable interest in critical phenomena associated with magnetic ordering. Most of the attention has focused on static properties such

as susceptibility or relatively simple dynamical properties such as spin diffusion coefficient. Our concern has been with the more complex problem of magnetic resonance linewidth in the vicinity of the critical point. In Report C00-1488-2, we arrived at some general results for behavior of linewidths in the critical region in the absence of an applied field and considering only the most divergent effects. These ideas have been used and acknowledged by R. M. White in his new text "Quantum Theory of Magnetism" (McGraw Hill, 1970). The Ph.D. thesis of P. Leichner calculated moments of the resonance line in the actual situation where one has an applied field. His work showed that divergent effects are removed, that examination of the moments can lead to a reasonable description of the linewidth in the critical region, and that - even in the critical region - short range order dominates the temperature dependence. These facts had not been appreciated before, and his work probably represents the first realistic attempt at calculating linewidth in sufficient detail that meaningful comparison can be made with experiment.

3. Nuclear Spin Waves (Reports 10, 14, 20; Publications 5, 8)

Theoretical as well as experimental work has been done on nuclear spin waves. We showed that nuclear spin waves alone could in principle be excited by parallel pumping in a flopped antiferromagnet. Previous workers had overlooked indirect coupling of the r-f field to the nuclei through the electronic spins and the high ellipticity of electronic precessional motion and thus came to the erroneous conclusion that nuclear spin waves by themselves could not be excited with reasonable r-f powers.

Our work on the theory of nuclear spin wave relaxation explained why NMR lines in antiferromagnets tend to be much narrower than originally predicted by theory and gave the first quantitative calculation for temperature and wave vector dependence of nuclear spin wave relaxation rates. Since the lifetimes calculated in this manner are orders of magnitude longer than at first suspected on basis of the Suhl-Nakamura interaction, we also have indicated how nuclear spin waves can be so well-defined.

4. Other Theories of Relaxation (Reports 12, 15; Publications 7, 9)

In collaboration with J. W. Culvahouse we derived for the first time an expression for paramagnetic resonance linewidth under conditions that the broadening is due to Orbach spin-lattice relaxation processes. The work strongly suggests the importance of bottlenecking even at very low concentrations of paramagnetic ions.

Relaxation in an Ising dipolar model was formulated and results compared with experiments on dysprosium ethyl sulfate (DyES). We showed that what had appeared to be anomalously short relaxation times could in fact be explained by reasonable values of spin-spin interactions in DyES.

List of Technical Reports

C00-1488-1. Progress Report

2. "On ESR Linewidths Near the Critical Temperature".
3. "Magnon-Magnon Interactions as Deduced from Harmonic Generation in YIG" (also publication 1).
4. "Time Correlations of Spin Operators in Finite Linear Magnetic Chains" (Ph.D. thesis, F. Carboni).
5. "Time Correlations and Magnetic Resonance Linewidth in Finite Heisenberg Linear Chains" (also publication 2).
6. "Exchange and the '10/3 Effect' in $\text{Cu}(\text{NH}_3)_4\text{SO}_4\cdot\text{H}_2\text{O}$ (also publication 3)
7. "Excitation of Nuclear and Electronic Spin Waves in RbMnF_3 by Parallel Pumping" (also publication 4).
8. Progress Report
9. "Interactions between Parametrically Excited Spin Waves in Yttrium Iron Garnet" (Ph.D. thesis, J. Bierlein).
10. "Nuclear Spin Wave Relaxation and Narrowing of NMR Lines in Ferro- and Antiferromagnets" (also publication 5).
11. "Time Dependence of Spin Operators in Finite Heisenberg Linear Chains" (also publication 6).
12. " T_1 and T_2 for Orbach Relaxation Processes" (also publication 7).
13. Progress Report.
14. "Excitation of Electronic and Nuclear Spin Waves in a Flopped Antiferromagnet" (also publication 8).

15. "Theory of Magnetic Relaxation in a Near-Ising System: DyES"
(also publication 9).
16. "Temperature Dependence of the Ferromagnetic Resonance Linewidth"
(Ph.D. thesis, P. K. Leichner).
17. "Direct Excitation of Two Nuclear Spin Waves by Parallel Pumping
in CsMnF_3 " (also publication 10).
18. "Harmonic Generation and Parametrically Coupled Spin Waves
in YIG" (also publication 11).
19. "Theory of Hyperfine ESR Linewidth in a Linear Chain:
 $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ " .
20. "Investigation of Nuclear and Electronic Spin Waves by Parallel
Pumping" (Ph.D. thesis, L. Hinderks).

Publications

1. Bierlein, J. D. Hinderks, L. W., and Richards, P. M., "Magnon-Magnon Interactions as Deduced from Harmonic Generation in YIG," J. Appl. Phys. 38, 1232 (1967).
2. Carboni, F. and Richards, P. M., "Time Correlation and Magnetic Resonance Linewidth in Finite Heisenberg Linear Chains," J. Appl. Phys. 39, 967 (1968).
3. Rogers, R. N., Carboni, F., and Richards, P. M., "Exchange and the '10/3 Effect in $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ '," Phys. Rev. Letters 19, 1016 (1967).
4. Hinderks, L. W. and Richards, P. M., "Excitation of Electronic and Nuclear Spinwaves in RbMnF_2 by Parallel Pumping," J. Appl. Phys. 39, 824 (1968).
5. Richards, P. M., "Nuclear Spin Wave Relaxation and Narrowing of NMR Lines in Ferro- and Antiferromagnets," Phys. Rev. 173, 581 (1968).
6. Carboni, F. and Richards, P. M., "Time Dependence of Spin Operators in Finite Heisenberg Linear Chains," Phys. Rev. 177, 889 (1969).
7. Culvahouse, J. W. and Richards, P. M., " T_1 and T_2 in Orbach Relaxation Processes," Phys. Rev. 178, 485 (1969).
8. Hinderks, L. W. and Richards, P. M., "Excitation of Electronic and Nuclear Spin Waves in a Flopped Antiferromagnet," Phys. Rev. 183, 575 (1969).
9. Richards, P. M., "Theory of Magnetic Relaxation in a Near-Ising System: DyES," Phys. Rev. 187, 690 (1969).
10. Adams, B. T., Hinderks, L. W., and Richards, P. M., "Direct Excitation of Two Nuclear Spin Waves in CsMnF_3 ," J. Appl. Phys. 41, 931 (1970).
11. Bierlein, J. D., and Richards, P. M., "Harmonic Generation and Parametrically Coupled Spin Waves in YIG," Phys. Rev. June, 1970.

Contributed Papers at American Physical Society Meetings and Magnetism Conferences.

1. "Magnon-Magnon Interactions as Deduced from Harmonic Generation in YIG", presented by P. M. Richards at 12th Annual Magnetism Conference, Washington, D. C., November, 1966. (Same as publication 1)
2. "Time Correlations and Magnetic Resonance Linewidth in Finite Heisenberg Linear Chains", presented by P. M. Richards at International Congress on Magnetism, Boston, September, 1967. (Same as publication 2)
3. "Excitation of Nuclear and Electronic Spin Waves in RbMnF_3 by Parallel Pumping", presented by L. W. Hinderks at International Congress on Magnetism, Boston, September, 1967. (Same as publication 4)
4. "Temperature Dependence of the Ferromagnetic Resonance Linewidth", Bull. Am. Phys. Soc. 14, 194 (1969); presented by P. K. Lechner at APS Meeting, St. Louis, February, 1969.
5. "Theory of Magnetic Relaxation in a near-Ising System: DyES", Bull. Am. Phys. Soc. 14, 408 (1969); presented by P. M. Richards at APS Meeting, Philadelphia, March, 1969.
6. "Direct Excitation of Two Nuclear Spin Waves by Parallel Pumping in CaMnF_3 ", presented by L. W. Hinderks at 15th Annual Magnetism Conference, Philadelphia, November, 1969. (Same as publication 10)
7. "Theory of Hyperfine ESR Linewidth in the Linear Chain = $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ ", Bull. Am. Phys. Soc. 15, 269 (1970); presented by C. McElwee at APS Meeting, Dallas, March, 1970.