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Paramagnetic spin fluctuations: a mode coupling interpretation of neutron scattering data for EuO, Pd_2MnSn and Fe

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Abstract. Spectra of paramagnetic spin fluctuations in insulating and metallic ferromagnets, obtained by inelastic neutron scattering, are interpreted in terms of appropriate Heisenberg models. A mode coupling approximation is used which takes full account of the lattice structure and the extended range of the exchange interaction, the form of which is obtained from the interpretation of spin wave spectra measured in the ordered state. The favourable outcome of confronting theoretical spectra with experimental data for EuO, Pd_2MnSn and Fe leads us to conclude that the mode coupling theory provides a more than adequate description of paramagnetic spin fluctuations. The dependence of the spectra on an increasing neutron wave vector is a change from a Lorentzian shape to a distinctly squared-up shape, but there is no strong evidence in any of the three systems of a damped collective mode contribution even for zone boundary wavevectors.

1. Introduction

In recent years a number of papers have reported results of neutron scattering experiments on isotropic ferromagnets in the paramagnetic phase. The availability of stronger neutron sources, and the development of new experimental techniques have made possible high resolution magnetic neutron scattering experiments for a large range of energies and wavevectors. The improvement in the quality of the experimental data allowed us to distinguish between the different theoretical predictions and to increase our knowledge about interactions in magnetic materials. Moreover, distinctive features of each material can be resolved experimentally. This calls for the formulation of better theories or the refinement of existing ones, since many of the latter were originally developed to interpret the general properties of ferromagnetic systems common to all such materials. The resurgence of theoretical studies on the dynamics of paramagnetic spin fluctuations has been prompted by debate on the features displayed in the new experimental data.

Amongst various ferromagnetic materials, those most frequently used in neutron scattering experiments are the europium compounds EuO and EuS, the Heusler compound Pd_2MnSn and the 3d metals Fe and Ni. The latter are typical examples of all ferromagnetic materials: in these elements, ferromagnetism was discovered and initially studied, and they are the basic components of materials commonly used in technical applications. On the other hand, the europium compounds are better physical realisations of localised moment ferromagnets (Passell *et al* 1976) and afford an important opportunity to test theories of ferromagnetism. Neutron scattering data on the Heusler

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compound Pd_2MnSn (Noda and Ishikawa 1976a, b) at a low temperature appear to be well described by a Heisenberg Hamiltonian.

Much theoretical work has been done on the nature of the ferromagnetic state, the origin of the magnetic interactions and the properties of spin waves in the ordered phase. The result of such theoretical efforts has been a satisfactory description of the magnetic ground state and thermal properties of the spin excitations. This has been achieved essentially within a perturbative framework for the Heisenberg model of a ferromagnet. No such framework exists for the paramagnetic phase since a small parameter has not been identified. At the critical point the renormalisation group technique provides a rigorous prescription of both static and dynamic events. However, the results cannot be unambiguously extended to the paramagnetic state, which is the central topic of this paper.

Neutron scattering experiments allow us to measure the response function $S(\boldsymbol{q}, \omega)$, which is connected to the Fourier transform $F_{\boldsymbol{q}}(\omega)$ of the normalised relaxation function (Lovesey 1987) by the fluctuation-dissipation theorem

$$S(\boldsymbol{q}, \omega) = \frac{\omega}{1 - exp(-\omega/k_B T)} \chi_{\boldsymbol{q}} F_{\boldsymbol{q}}(\omega) \tag{1}$$

where $\chi_{\boldsymbol{q}}$ is the static susceptibility.

Paramagnetic neutron scattering data are often interpreted in terms of the so-called 'three-pole approximation' (Lovesey and Meserve 1972). This approximation is obtained from a truncation of a continued fraction expansion of the Laplace transform of the spin relaxation function, namely

$$F_{\boldsymbol{q}}(\omega) = \frac{1}{\pi} \frac{\tau_{\boldsymbol{q}} \delta_{1\boldsymbol{q}} \delta_{2\boldsymbol{q}}}{[\omega \tau_{\boldsymbol{q}} (\omega^2 - \delta_{1\boldsymbol{q}} - \delta_{2\boldsymbol{q}})]^2 + (\omega^2 - \delta_{2\boldsymbol{q}})^2} . \tag{2}$$

Here, $\boldsymbol{q}$ is a wavevector within a magnetic Brillouin zone. The integral of this expression for $F_{\boldsymbol{q}}(\omega)$ is unity. Integrals of the expression times ω^2 and ω^4 generate simple relations, which are independent of τ, between δ_1 and δ_2 and the second and fourth frequency moments. The quantities δ_1, δ_2 appearing in equation (2) are functions of static temperature dependent spin correlation functions and can in principle be evaluated from the Hamiltonian and known exchange interaction constants. In contrast the time $\tau_{\boldsymbol{q}}$ is not well defined because it depends on the criterion adopted for the truncation of the continued fraction expansion. For example it is possible to argue (Lovesey and Meserve 1972) that $\tau_{\boldsymbol{q}}$ can be expressed in terms of $\delta_{2\boldsymbol{q}}$. Although the three-pole approximation has proved to be very useful for interpreting experimental data (Böni and Shirane 1986, Bohn *et al* 1984), it is not really satisfactory, because it is phenomenological — and it is not clear how and up to which point it can account for the dynamical coupling among the various modes of fluctuation. Certainly, within the three-pole approximation it is not possible to obtain the correct dynamical critical behaviour in which the conserved modes play a dominant role.

An approach that is able to give, self-consistently, the static quantities involved in the Mori expansion for the relaxation function has been developed by Lindgård (1983). His theory is essentially a moment expansion based on a two-pole approximation for $F_{\boldsymbol{q}}(\omega)$, and its application to europium compounds shows that it is not entirely able to describe the experimental data for large wavevectors, expecially near and at T_c.

A more satisfactory approach to the study of spin fluctuations in the paramagnetic phase is the mode-coupling theory (Hubbard 1971, Kawasaki 1975). This non perturbative theory has been developed, over a period of many years, to treat disordered states of matter in a variety of different physical systems.

The merit of mode-coupling theory is that the underlying approximations are reasonable in the critical and paramagnetic phases, and for all wavevectors. It not only gives the correct dynamical critical behaviour for an isotropic Heisenberg ferromagnet, but it also explains the line-shape modifications at very small q, observed in neutron spin-echo experiments, due to the onset of the dipolar interaction (Frey and Schwabl 1988, Frey *et al* 1988).

The use of the mode-coupling theory to interpret neutron scattering data collected out of the critical region, i.e. for high wavevectors and temperatures, was up to now prevented by the fact that the known solutions of the integro-differential equations for the relaxation function are obtained either analytically, for asymptotically small q, (Lovesey and Williams 1986) or numerically, but only for a simple cubic lattice with magnetic interactions restricted to nearest neighbours (Hubbard 1971).

In a previous paper (Cuccoli *et al* 1989) we have shown that the results of magnetic neutron scattering experiments on europium compounds are well reproduced by the numerical solution of the equations for an FCC lattice allowing for interactions with two sets of neighbours. An aim of this paper is to explore further the capability of the mode-coupling theory to interpret paramagnetic neutron scattering data. This is achieved by using the theory to interpret new high resolution experimental data on europium oxide, and data for other compounds that possess longer range interactions and different cubic lattice structures.

The equation for the relaxation function of a Heisenberg magnet in the mode-coupling approximation is reviewed in section 2. Results appropriate for EuO, Pd_2MnSn and Fe are presented in section 3, together with comparisons between theoretical and experimental results. Our conclusions are gathered in section 4.

2. Theory

Application of the mode-coupling approximation to the study of the spin fluctuations in a Heisenberg magnet leads to the following equation for the relaxation function:

$$\dot{F}_q(t) = -2T \sum_k (J_k - J_{q-k})(J_k - J_q)\chi_k \int_0^t \mathrm{d}t' \, F_k(t-t')F_{q-k}(t-t')F_q(t') \tag{3}$$

where χ_q is the static susceptibility and J_q the Fourier transform of the exchange interaction.

The numerical method employed to solve equation (3) has been described in detail by Cuccoli *et al* (1989). We recall only that, for the static susceptibility, which must be inserted in the theory, we have used the spherical model, because the form is consistent with the dynamical equations. Moreover this susceptibility is in satisfactory agreement with experimental data if the temperature is not too close to T_c.

The need to have a prescription for the susceptibility is a weakness of the mode-coupling theory, since the prescription is necessarily approximate. We do not know how the uncertainty in susceptibility influences the usefulness of the theory. A similar

dilemma exists in using the three-pole approximation. For although we have exact expressions for δ_1 and δ_2 in terms of static correlation functions we do not have exact values for them. The approximate method used in the past to compute δ_1, δ_2 (Young and Shastry 1982) is similar in spirit to our use of the spherical model. Perhaps the only really satisfactory route is to compute the susceptibility and correlation functions in δ_1, δ_2 by computer simulation of a Heisenberg model with appropriate exchange integrals.

3. Interpretation of data

3.1. EuO

The solution of equation (3) obtained for EuO has been successfully compared by Cuccoli *et al* (1989) with the experimental data collected at Brookhaven by Böni and Shirane (1986). The comparison is made with their fitted function to take some account of the resolution effects. We now have experimental work on EuO reported by Mezei *et al* (1989) with a very high energy resolution which allows a direct comparison of theoretical results with the experimental data. Such a comparison with the solution of the mode-coupling equation (3) is displayed in figure 1. We also conclude that in this case good agreement is reached. Note in figure 1(*b*), that the broken curve represents

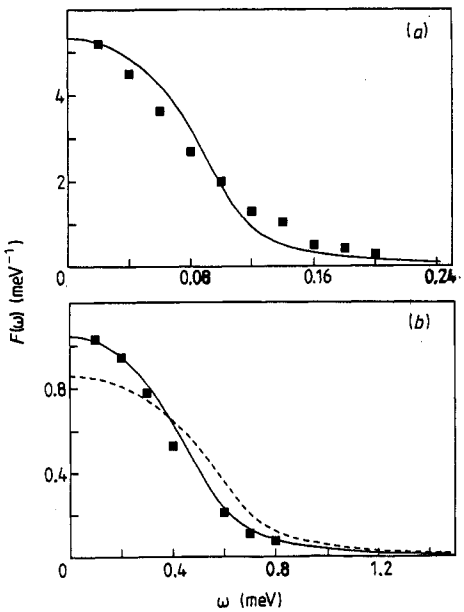

Figure 1. Experimental data reported by Mezei *et al* (1989) for EuO at its critical temperature are compared with results of the mode-coupling approximation for (*a*) $q = 0.15$ Å^{-1} and (*b*) $q = 0.3$ Å^{-1}. The exchange constants used, and other details of the calculation, are the same as those used by Cuccoli *et al* (1989). The broken curve for $q = 0.3$ Å^{-1} is the result of the mode-coupling calculation for a simple cubic lattice, and it demonstrates the influence of the lattice structure on the dynamic properties even at relatively long wavelengths.

the solution of equation (3) for a SC lattice with nearest neighbour interactions only. It is seen that for wavevectors that are not too small the use of the true lattice structure and all exchange constants is of fundamental importance if we wish to reproduce the experimental data very well without any adjustable parameters.

3.2. Pd_2MnSn

An experimental investigation of the dynamics of spin fluctuations in Pd_2MnSn for intermediate and large wavevectors has been reported by Graf *et al* (1989). Experimental work devoted to spectra near or at the zone boundary is of particular relevance, because it constitutes an important test of current microscopic descriptions of ferromagnetic systems. Moreover it is at high momentum transfer that there are most doubts in our knowledge of the statistical properties of paramagnetic spin fluctuations, since for small q the conservation equation dictates the form of the spin correlation function, in the paramagnetic region.

Pd_2MnSn is similar in some respects to EuO , in as much as it is a FCC ferromagnet with a relatively low critical temperature ($T_c \simeq 193$ K), but it is metallic. Nevertheless, in the ordered phase, the temperature dependence of the spin waves energies and damping is well described by the interacting spin wave theory for a Heisenberg magnet, if allowance is made for interactions up to the eight shells of neighbours, which show an oscillating character at large distances. Therefore it has been suggested (Noda and Ishikawa 1976b) that Pd_2MnSn can be considered an ideal Heisenberg isotropic ferromagnet with long range interactions.

It is interesting to enquire whether a Heisenberg model for Pd_2MnSn , with the exchange constants fixed to the value obtained from the fitting of the spin wave dispersion curves, is also able to interpret the paramagnetic spectra, as in the case of europium compounds. For the latter, results from the three-pole approximation and the mode-coupling theory generally agree with each other and with the experimental data. The only significant difference between the three sets of results occurs for large wavevectors. Well defined inelastic shoulders for large wavevectors are generated by the three-pole approximation, but these are only weakly present in the solution of mode-coupling equations. They are more evident in the experimental data for EuS than in the data for EuO, but even in EuS the inelastic shoulders are not a striking feature.

Graf *et al* (1989) analyse the available experimental data on Pd_2MnSn at $T = 1.1\,T_c$ in terms of the three-pole approximation. It has been argued that a good estimate for the time τ in equation (2) is $\tau = (2/\pi\delta_2)^{1/2}$. Using this expression the relaxation function is completely determined by the two quantities δ_1 and δ_2. Treating these as fitting parameters, the three-pole approximation turns out to be a good functional form to reproduce experimental line shapes.

The physical significance of these results is enhanced by the fact that, the authors, find the values of δ_1 and δ_2 obtained by fitting are in qualitative agreement with those obtained from an approximate evaluation of the molecular formulae for δ_1 and δ_2 derived from their relation to frequency moments of the spectrum. We conclude that, the three pole approximation is useful for the interpretation of paramagnetic spectra and that values of δ_1 and δ_2 required to obtain good fits are physically realistic. The authors find the spectra obtained from the three-pole approximation using calculated values of δ_1, δ_2 are inferior to the fits. The deterioration of the spectra is most pronounced at high frequencies. This is readily understood since δ_1, δ_2 are related to frequency moments which, of course, relate most strongly to the high frequency

features of the spectra. We note that the difference between calculated and fitted parameters is more pronounced for δ_2, which is also related to the determination of τ.

The occurrence of a high frequency peak in the spectra can be related to the values of δ_1, δ_2. Using either the three-pole approximation (a form of Padé approximation) or an expansion in terms of Hermite polynomials it is readily shown that a high frequency peak exists in a spectrum if the quantity (Lovesey 1987)

$$\alpha = \frac{1}{3}\left(\frac{\delta_2}{\delta_1} - 2\right)$$

is negative. It is easy to check that $\alpha = 0, \infty$ for Gaussian and Lorentzian function, respectively. Values of α calculated using δ_1, δ_2 obtained from fits to experimental data are positive, and decrease by more than a factor of five on going from the smallest to the largest wavevector. These findings are consistent with a squaring of the spectrum with increasing wavevector.

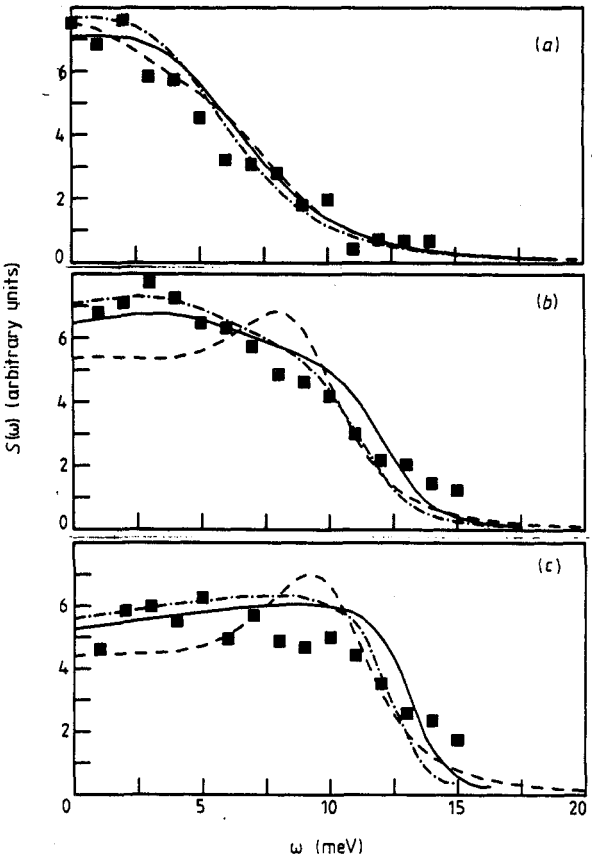

Figure 2. Experimental data (Graf *et al* 1989) for the Heusler compound Pd_2MnSn at $T = 1.1\,T_c$ and three wavevectors in the (111) direction are shown together with results from theoretical calculations: (*a*) $q = 0.5\,q_{ZB}$, (*b*) $q = 0.7\,q_{ZB}$, (*c*) $q = 0.92\,q_{ZB}$. The full and chain curves are mode-coupling calculations with six and eight shells of neighbours, respectively, and the broken curve is the theoretical three-pole approximation. Calculated spectra are convolved with a resolution function, as described in the text.

We have obtained the solution of the mode coupling equation to compare with Pd_2MnSn data by making use of the exchange constants given by Noda and Ishikawa (1976a) and setting $S = 2.1$, which is the value deduced from the observed magnetic moment. The mode coupling numerical calculation has been executed both using only the first six exchange constants, as done by Graf *et al* (1989) to compute the theoretical value of δ_1 and δ_2, and all the eight constants given by Noda and Ishikawa (1976a). The theoretical relaxation function is multiplied by the detailed balance factor (1) and, to take some account of the spectrometer resolution, we have convoluted it with a Lorentzian having the quoted experimental half width. The normalisation constant has been fixed from the fitted three-pole function.

The results of the comparison are shown in figure 2, where the mode coupling solution is reported together with the experimental data and the theoretical three-pole function with δ_1 and δ_2 evaluated by Graf *et al* (1989). The mode coupling solution is superior to the three-pole approximation. Particularly good agreement between theory and data is obtained for $q = 0.5q_{ZB}$ and $q = 0.70q_{ZB}$, while for $q = 0.92q_{ZB}$ the improvement with respect to the three-pole approximation is not meaningful ($q_{ZB} = (0.5, 0.5, 0.5)2\pi/a$, where a is the lattice constant and q_{ZB} is the zone boundary wavevector along the (111) direction). New experiments on Pd_2MnSn devoted to other directions of the wavevector, and convolution of the mode coupling results with the resolution function, are important factors in a further study. Even so, we believe our first attempt demonstrates the adequacy of the mode coupling theory to interpret Pd_2MnSn data, and that it is superior to the three-pole approximation.

Table 1. Set of exchange constants proposed to model iron with a Heisenberg Hamiltonian. $T_c^{MF} = \frac{1}{3}S(S+1)J_0$ e T_c^{SM} are, respectively, the mean field and spherical model calculated critical temperature; D is the calculated spin-wave stiffness constant.

	I[a]	II[b]	III[c]
J_1 (K)	114.6	390.7	51.7
J_2 (K)	161.9	113.2	56.2
J_3 (K)	145.1	−52.9	−2.9
J_4 (K)	—	—	46.4
J_5 (K)	−132.1	—	−13.3
J_6 (K)	—	—	5.2
J_7 (K)	—	—	1.7
J_8 (K)	—	—	1.7
J_9 (K)	—	—	−1.7
J_{10} (K)	—	—	18.0
T_c^{MF} (K)	1715	2113	1293
T_c^{SM} (K)	1282	1385	1049
D (meV Å^2)	236	211	549

[a] Shastry *et al* (1981).
[b] Johnson *et al* (1987).
[c] Wang *et al* (1982).

3.3. Fe

Iron is a metallic BCC ferromagnet which owes its magnetic properties to 3d electrons, and itinerant effects are surely important. Nevertheless attempts have been made to interpret experimental data on iron in terms of a localised spin model (Shastry *et al*

1981, Johnson *et al* 1987), deriving the exchange constants from spin wave dispersion curves. The disagreement found between the predictions of the Heisenberg model and the experiments was sufficient for Johnson *et al* (1987) to conclude that the model is not physically meaningful. We think that such a conclusion is too hasty. First, dynamical behaviour has been analysed only within the three-pole approximation and, as shown in Pd_2MnSn this is not entirely satisfactory. Secondly, the exchange interaction has been artificially restricted. The latter influences the high-q behaviour much more than the low-q response. Moreover theoretical calculations by Wang *et al* (1982) have shown that the free energy of the magnetic excitation can be mapped into an effective Heisenberg Hamiltonian only if long range interactions are allowed.

In view of these misgivings we have computed the solution of the mode coupling equations for a BCC lattice, with $S = 1$, using the three sets of exchange parameters given in table 1. Results of the calculation for $T = 1.28\,T_c$ are shown in figure 3.

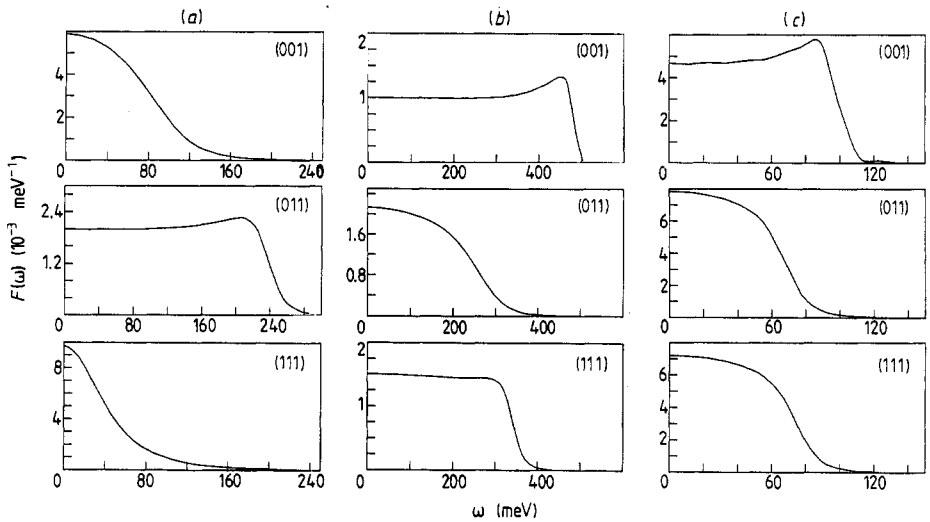

Figure 3. Paramagnetic spectra for Fe calculated from the mode-coupling equation for three different sets of exchange constants, listed in table 1. The temperature $T = 1.28\,T_c$ and the wavevectors are at the zone boundaries in the (001), (011) and (111) directions. Parts (*a*), (*b*) and (*c*) refer, respectively, to the first, second and third set of exchange constants.

The three sets of exchange constants are listed in table 1, together with the mean field and spherical model critical temperatures, and the spin wave stiffness constants. The measured values are $T_c = 1043$ K and $D = 0.28$ eV Å^2. By comparing values for T_c and D computed from the exchange constants with measured values we find that no set of exchange constants is outstandingly good.

Turning to the prediction of the coupled mode theory, displayed in figure 3, it is at once apparent that the different exchange constants give significantly different spectra. Moreover it is interesting to note the strong dependence of the width of the spectra at the zone boundary on the direction of the wavevector. The simple relation between the magnitude of the exchange constants and the spectral widths based on an isotropic behaviour of the system is not valid.

With the limited available constant-q experimental data on iron (Brown *et al* 1985, Shirane *et al* 1986) it is not meaningful to attempt a detailed confrontation between

theory and data. However, the exchange constants III, which extend to ten shells of neighbours, seem to show a convergence and give a more realistic paramagnetic spectra. Spectra for zone boundary wavevectors are observed to decrease in magnitude quite rapidly beyond 100 meV, which is consistent with our calculation for the set of exchange constants III. For this set of constants, the shape of the spectrum does not change appreciably with wavevector in the second half of the range in the (011) and (111) directions. This result is in qualitative agreement with the experimental observation in the (011) direction (Shirane *et al* 1986).

4. Conclusions

The mode coupling approximation, for the dynamic properties of paramagnetic spin fluctuations in Heisenberg magnets, appears to provide a good interpretation of experimental data in insulating and metallic magnets. Good agreement between calculations and data requires the use of the correct lattice structure, and exchange constants. Spectra are found to be sensitive to the precise form of the exchange interaction, and this sensitivity can be exploited to distinguish between interactions that give tolerable spin wave dispersions, critical temperatures and other measurable quantities.

This finding adds more weight to the value of inelastic neutron scattering measurements at the critical temperature and in the paramagnetic phase. Certainly, it seems that additional high resolution and constant-q data for paramagnetic Fe, and its interpretation in terms of a coupled mode theory, would go a long way toward establishing the nature of the magnetic interactions.

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