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INTRODUCTION TO NEUTRON SCATTERING BY MAGNETIC EXCITATIONS*

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I feel that magnetism is one of the most interesting and challenging fields in condensed matter physics. There are a wide variety of magnetic phenomena and measurements provide a test of the many-body theories that are necessary to understand condensed matter physics of all types. Indeed the famous theorem quoted by Van Vleck[1] as "Miss van Leeuwen's theorem" tells us that classical statistics applied to any metallic system results in no magnetism so that quantum mechanics is necessary to even get started. The magnetic moment of neutrons makes them an excellent probe of magnetic phenomena and very direct information is obtainable with neutrons. In fact the generalized susceptibility is directly given by a neutron scattering experiment, and this quantity tells all there is to know about a magnetic system. The purpose of this paper is to serve as an introduction to a series of papers on magnetic excitations. I will thus establish the necessary neutron scattering formalism so that it will not have to be repeated by each author and discuss briefly various types of magnetic systems. The symbols used will be consistent with those used by Marshall and Lovesey.[2] Extensions and full discriptions of some of the cross section derivations can be found in their book.

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The magnetic field caused by an electron moving with velocity v_e is

$$H = \mathrm{curl}\left(\frac{\bar{\mu}_e \times \bar{R}}{|\bar{R}^3|}\right) + \frac{(-e)}{c} \quad \frac{\bar{v}_e \times \bar{R}}{|\bar{R}^3|} \tag{1}$$

We can thus define an interaction potential between the neutron and the electron by

$$- \gamma \, \mu_N \, \hat{\sigma} \cdot \bar{H} \tag{2}$$

where 1/2 $\hat{\sigma}$ is the spin of the neutron, μ_N is the nuclear magneton and $\gamma = -1.91$. The neutron cross section in the Born approximation is given by

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k} \left(\frac{m}{2\pi h^2}\right)^2 \sum_{\lambda\delta} P_\lambda \, P_\sigma \sum_{\lambda'\sigma'} |\langle k'\sigma'\lambda'|V|k\sigma\lambda\rangle|^2$$

$$\times \; \delta(\text{Energy}) \tag{3}$$

where k and k' are the incoming and outgoing neutron wave vectors, λ and λ' denote the quantum numbers required to specify the initial and final states of the target and P_λ and P_σ are the initial state probabilities.

If we insert the potential given in (2) into the cross section formula (3) and perform some algebra, we arrive at

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k} \sum_{\lambda\lambda'} P_\lambda \, \langle\lambda|\bar{Q}_\perp^+|\lambda'\rangle\langle\lambda'|\bar{Q}_\perp|\lambda\rangle$$

$$\times \; \delta(\text{Energy}) \tag{4}$$

where $Q_\perp$ is the total magnetic interaction operator. The cross section is often written in terms of a similar operator $\hat{Q}$ defined by $\bar{Q}_\perp = \bar{K}x(\hat{Q} \times \bar{K})$, $\bar{K}$ being the scattering vector $\bar{k} - \bar{k}'$. We then obtain

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \hat{K}_\alpha \hat{K}_\beta) \times$$

$$\sum_{\lambda\lambda'} P_\lambda \langle\lambda|\hat{Q}_\alpha|\lambda'\rangle\langle\lambda'|\hat{Q}_\beta|\lambda\rangle \times \delta(\text{Energy}) \qquad (5)$$

α and β being the Cartesian component index.

Perhaps the easiest magnetic system to deal with is one in which a crystalline field splits a ground state degeneracy into a series of discrete levels. If the exchange interaction is weak, it is sufficient to consider the cross section for scattering by a single isolated ion. If the crystalline field has cubic symmetry and we let Γ_n be the irreducible representations of the cubic group, the cross section for a transition $\Gamma_n \to \Gamma_n'$ is given by

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 \frac{k'}{k} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \hat{K}_\alpha \hat{K}_\beta) \times$$

$$\sum_{\nu,\nu'} P_n \langle\Gamma_{n\nu}|\hat{Q}_\alpha^+|\Gamma_{n'\nu'}\rangle\langle\Gamma_{n'\nu'}|\hat{Q}_\beta|\Gamma_{n\nu}\rangle \times \delta(\text{Energy}) \qquad (6)$$

where we have denoted the wavefunctions by $|\Gamma_{n\nu}\rangle$ where ν distinguishes the degenerate wavefunctions. If we only consider dipole transitions, $\hat{Q} = 1/2\ g\ F(k)\hat{J}$, where g is the gyromagnetic ratio, F(k) the magnetic form factor or the Fourier transform of the magnetic spin density, and $\hat{J}$ the total angular momentum spin operator. Conveniently for us, Birgeneau[3] has worked out the cross section for J values of interest, and graphs are provided for each J giving transition probabilities in terms of the Lea, Leask, and Wolf parameter w. The number w is just the ratio of the fourth and sixth degree terms of the Stevens operator equivalents.

Crystal field spectroscopy has now been performed at a number of laboratories. Time-of-flight is an excellent technique for making the measurements since data can be taken at all $\vec{K}$. The high energy neutrons available from the pulsed source may enable new materials to be studied, especially transition metal compounds where the crystal field splitting may be near the electron volt range. Certain of the actinides may also display crystal field transitions at high energies, the best cases probably being the heavier materials of the series.

As the exchange between ions increases, the discrete crystal field levels will broaden and eventually magnetic long-range order will occur. The simple crystal field description is then no longer satisfactory although many materials exist in the intermediate range where broad but still visible crystal field levels can be observed. Additional factors affect the magnetic ground state such as valence fluctuations in materials like SmS or some of the cerium compounds. In this case the magnetic scattering may consist of quasielastic scattering or rather diffuse scattering extending to high energy transfers. M. Lowenhaupt will discuss neutron spectroscopy from these types of systems.

There has always been a great deal of interest in the magnetism of the iron group transition metals, Fe, Ni, and Co. The crystal field effects are large in these materials so that L,S coupling is broken down, and since the expectation value of L for any nondegenerate state is 0, only spin needs to be considered to first order. Spin-orbit coupling results in some orbital contribution to the neutron cross section which is important at the higher momentum values, but I will neglect it in the following discussion.

We know the electrons responsible for the magnetism in Fe, Ni, and Co give a large contribution to the low temperature specific heat and that the moment values are nonintegral so that the electrons are at least partly itinerant in nature, and proper cross sections for neutron scattering should be developed from band theoretical considerations. The Hamiltonian for this case is of the form

$$H = \sum_i \frac{1}{2m} \bar{r}_i^2 + V(\bar{r}) + \sum_{i \neq j} \frac{e^2}{|\bar{r}_i - \bar{r}_j|} \tag{7}$$

where $\bar{p}$ and $\bar{r}$ are the momentum and position operator for the ith electrons and $V(\bar{r})$ is the periodic crystal potential from the ion cores. The first two terms consist of the kinetic energy of the electrons in the periodic potential of the crystal. The eigenfunctions of this part of the Hamiltonian are the block functions. The third term is the coulomb interaction of the electrons. One then assumes narrow bands and that the Wannier functions overlap from site to site is small. The Hubbard Hamiltonian can then be obtained and using second quantization formalism it is be given below

$$H = \sum_{k\sigma} E_k c^+_{k\sigma} c_{k\sigma} + 1/2\ I \sum_{i\sigma} c^+_{i\sigma} c^+_{i\sigma} c_{i\sigma} c_{i\sigma} \tag{8}$$

where σ represents the electron spin index. The first term represents the band energies and the second term the Coulomb interaction of electrons of opposite spin at the same site. From this the susceptibility can be calculated in the random phase approximation.

For the spin only case the magnetic interaction operator $\vec{Q}$ used in the cross section reduces to

$$\bar{Q} = \sum_{\ell} e^{i\bar{K}\cdot\bar{r}_\ell}\, \bar{S}_\ell \tag{9}$$

where the sum runs over the lattice sides ℓ of the crystal. In this case

$$\langle\lambda'|\bar{Q}|\lambda\rangle = \sum_{\ell} e^{iK\cdot r_\ell}\, F(\bar{K})\, \langle\lambda'|S_\ell|\lambda\rangle \tag{10}$$

It turns out to be convenient to include the energy δ function directly into the cross section. Using the integral representation of the δ function the cross section can be written in the form

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 1/2\; g(F(K))^2\, \frac{k'}{k} \sum_{\alpha,\beta}\left(\delta_{\alpha,\beta} - \hat{K}_\alpha \hat{K}_\beta\right) x$$

$$\frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt\; e^{-i\omega t} \langle S_K^\alpha(0)\; S_{-K}^\beta(t)\rangle \tag{11}$$

g being the gyromagnetic ratio.

Quite often it is useful to write the cross section in the form of the generalized susceptibility

$$\chi^{\alpha\beta}(\omega) = g\; \mu_B \frac{i}{\hbar} \int_{-\infty}^{\infty} dt\; e^{i\omega t} \langle[S^\alpha(t), S^\beta(0)]\rangle \tag{12}$$

The cross section then becomes

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 (1/2\ F|\bar{K}|)^2 \frac{k'}{k} \frac{N}{\pi\mu_\beta^2} \frac{e^{\hbar\omega\beta}}{e^{\hbar\omega\beta}-1}$$

$$\sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \hat{K}_\alpha \hat{K}_\beta) \text{ Im } \chi_{\bar{K}}^{\alpha\beta}(\omega) \tag{13}$$

where $1/\beta$ equals Boltzmann's constant times the temperature.

The result of a calculation of the transverse component of the susceptibility for the itinerant electron system is

$$\chi_{\bar{K}}(\omega) = \frac{\chi_{\bar{K}o}(\omega)}{1 - I/(g\mu_\beta)^2\ \chi_{\bar{K}o}(\omega)}\ , \tag{14}$$

where

$$\chi_{\bar{K}o}(\omega) = -\ (g\mu_\beta)^2 \frac{1}{N} \sum_{g} \frac{f_{\bar{K}+\bar{q}}\uparrow - f_{\bar{K}}\downarrow}{E_{\bar{K}+\bar{q}} - E_{\bar{q}} + \Delta + \hbar\omega + i\varepsilon}\ . \tag{15}$$

$\chi_{\bar{K}o}$ is generally called the noninteracting susceptibility. It contributes to the cross section when its denominator has zeros so that excitations fan out from an energy which is the splitting parameter for up and down spins. These are called single particle excitations. The zeros of the denominator of the interacting susceptibility give us an excitation that goes to zero energy as $\bar{K} \to 0$ and this is called the spin wave mode of the itinerate system. This is shown in Fig. 1 where the excitation spectrum consists of a spin wave which increases in energy from $\bar{K} = 0$ and rises to meet the single particle excitations that spread out from Δ.

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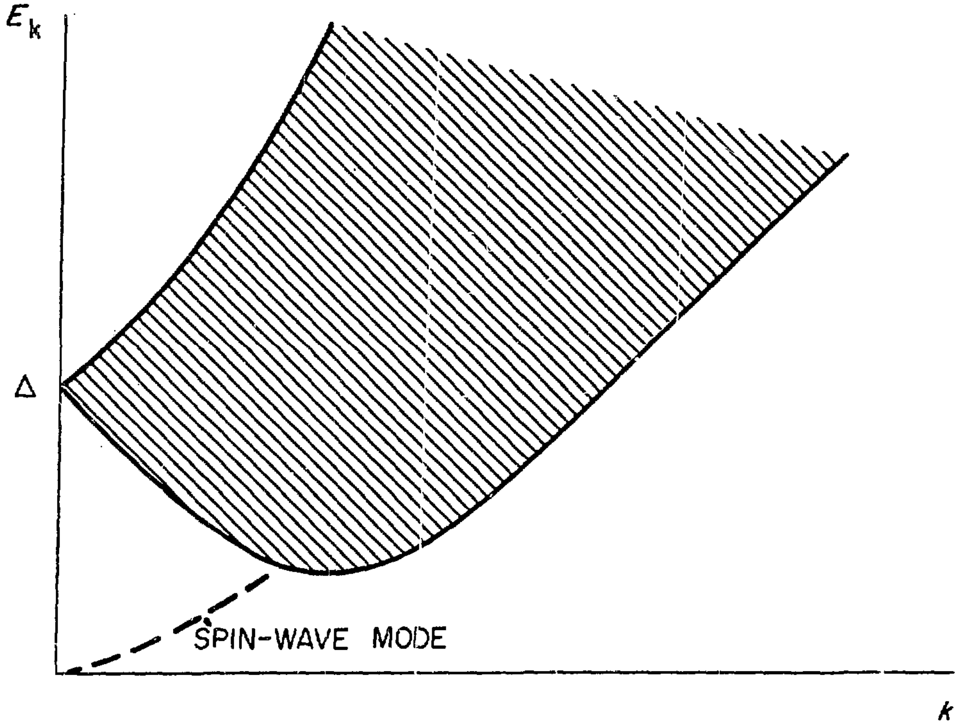


Fig. 1. Excitation spectrum expected for an itinerant magnet. Single particle excitations spread out from the spin splitting energy Δ. A spin wave mode rises from E = 0 at K = 0 and disappears as it enters the region of single particle excitations.

The most detailed measurements have been made on Ni. Figures 2 and 3 show contour maps of the measured scattering. The spin wave intensity clearly drops off rather rapidly with increasing energies for the [111] direction, and it appears that the spin wave is entering the region of single particle excitations around 25 THz (100 meV) at which point it becomes strongly damped and disappears. For the [100] direction the spin wave also weakens at higher energies but anomalous behavior is found at about 30 THz. This behavior is

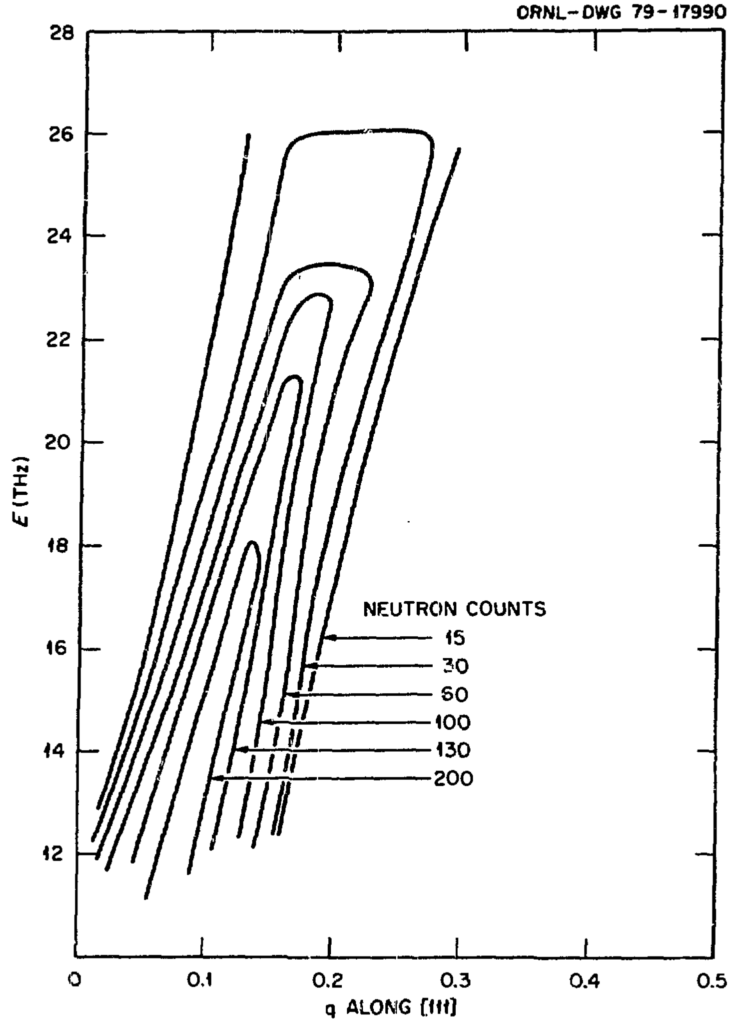


Fig. 2. Contour map of the scattering from the [111] direction for nickel at room temperature. The spin wave loses intensity rapidly as it enters a high density region of single particle excitations.

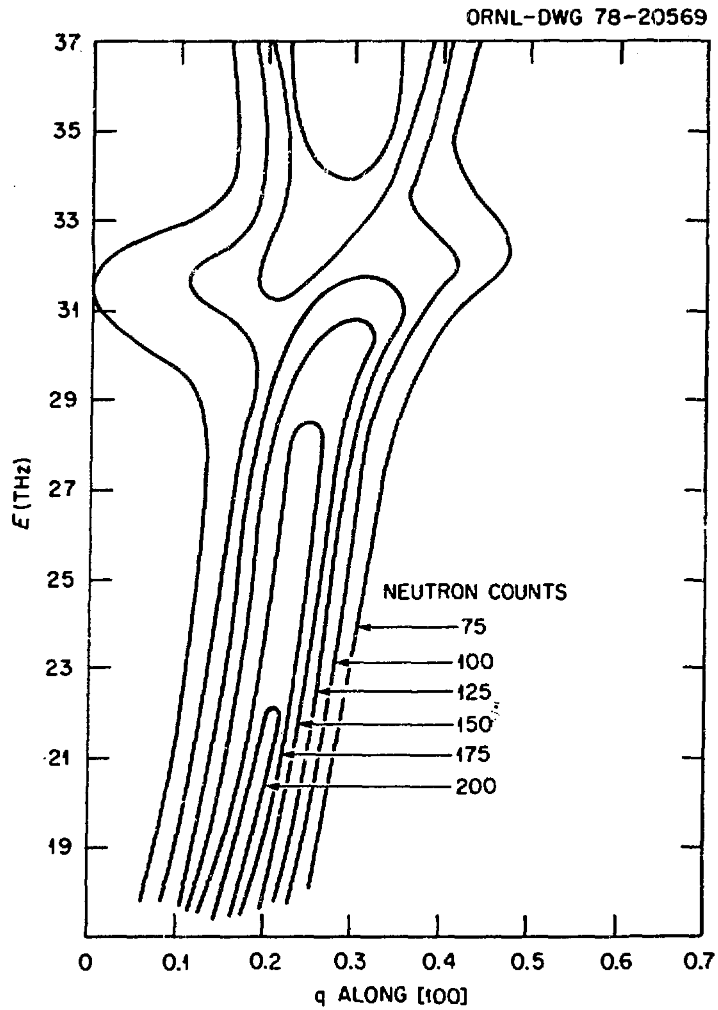


Fig. 3. Contour map of the scattering for the [100] direction for nickel. Structure near 31 THz ~ 120 meV suggests that an optical spin wave branch may cross the main acoustic spin wave branch.

in good agreement with a prediction by Cooke and Davis[4] that an optical spin wave should cross and interfere with the main spin wave branch at about this energy. Unfortunately the optical spin wave has not been seen directly despite repeated attempts. Calculations using the local density approximation by Callaway et al.[5] show no optical spin wave at 30 THz but do suggest the possibility of such a mode at a considerably higher energy. It would be very desirable to see the optical mode directly and perhaps the large number of epithermal neutrons produced by the spallation source would make this possible.

One can also write the susceptibility for longitudinal excitation of the itinerant electrons and the longitudinal noninteracting susceptibility is given by

$$\chi^{L}_{\bar{K}o}(\omega) = -\,1/2\; g\mu_{\beta}\,\frac{1}{N}\sum_{\sigma\bar{q}} \frac{f_{\bar{K}+\bar{q}o} - f_{\bar{q}o}}{E_{\bar{K}+\bar{q}} - E_{\bar{q}} + \hbar\omega + i\varepsilon} \tag{16}$$

The interacting susceptibility is given by an equation similar to (14). It is obvious that these excitations are nothing like spin wave excitations, and it is interesting to study their behavior. Shirane and Als-Neilsen[6] have apparently observed these excitations in iron for temperature slightly below T_C; however, additional experiments to further study their behavior would be useful.

Spin wave behavior is usually thought of in terms of a Heisenberg Hamiltonian with an external field along z.

$$H = -\sum_{\ell\ell'} J(\ell - \ell')\, S_\ell \cdot S_{\ell'} - g\mu_\beta \sum_\ell S^z_\ell \tag{17}$$

This can be written

$$H = -\sum_{\ell\ell'} J(\ell - \ell')\, S^z_\ell S^z_{\ell'} + S^+_\ell S^-_{\ell'} - g\mu_\beta H \sum_\ell S^z_\ell \tag{18}$$

and then one can solve the equation of motion for the operator S^+

$$ih\, \hat{S}^+_\ell = [\hat{S}^+_\ell, H] \tag{19}$$

which yields the spin wave energies

$$h\omega_{\bar{q}} = g\mu_\beta H + 2S(J(0) - \bar{J}(q))$$

$$J(\bar{q}) = \sum_\ell J(\ell)\, e^{i\bar{q}\cdot\bar{\ell}} \tag{20}$$

The cross section for spin wave creation is then

$$\frac{d^2\sigma}{d\Omega dE'} = \left(\frac{\gamma e^2}{mc^2}\right)^2 1/2\; g(F(\bar{K}))^2 \frac{k'}{k} (1 + \hat{K}^2_z)\; 1/2\; S$$

$$\frac{(2\pi)^3}{V_0} \sum_{\bar{q}\,\bar{\tau}} (n_{\bar{q}} + 1)\, \delta(\hbar\omega_{\bar{q}} - \hbar\omega)\, \delta(\bar{K} - \bar{q} - \bar{\tau}) \tag{21}$$

where $n_{\bar{q}}$ is the spin wave population factor, V_0 is the unit cell volume and τ is a reciprocal lattice vector. The longitudinal cross section in this case leads to no inelastic scattering.

The temperature dependence of magnetic excitations of ferromagnets especially near and above T_C is a topic of great recent interest. Figure 4 shows the spin wave spectra and transition temperatures of three ferromagnets. The maximum in the spin wave energy for EuO and Gd is equal to kT_C so that the ferromagnetic transition can be thought to be the result of exciting all spin wave modes. Clearly this is not the case for Ni and the nature of the transition in this material is of special interest.

In the region near and above T_C the cross section is often decomposed into the isothermal wave vector dependent susceptibility $\chi^{\alpha}_{\bar{K}}$ and a spectral weight function $F^{\alpha}(K,\omega)$. In this case the cross section for inelastic scattering is given by

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{\gamma e^2}{mc^2}\frac{k'}{k}\left(1/2\ gF(\bar{K})\right)^2 \sum_{\ell} \left(1 - \bar{K}_{\alpha}^2\right) S^{\alpha}(\bar{K},\omega)$$

where

$$S^{\alpha}(\bar{K},\omega) = \frac{N}{(q\mu_{\beta})^2}\ \chi^{\alpha}_{\bar{K}}\ \omega\ \frac{e^{\hbar\omega\beta}}{e^{\hbar\omega\beta}-1} F^{\alpha}(\bar{K},\omega)\ . \tag{22}$$

If one assumes an instantaneous spin correlation function of the form

$$\langle S_0^{\alpha}(0)\ S_{\ell}^{\alpha}(0)\rangle \sim \frac{e^{-\bar{K}\cdot\bar{R}}}{R} \tag{23}$$

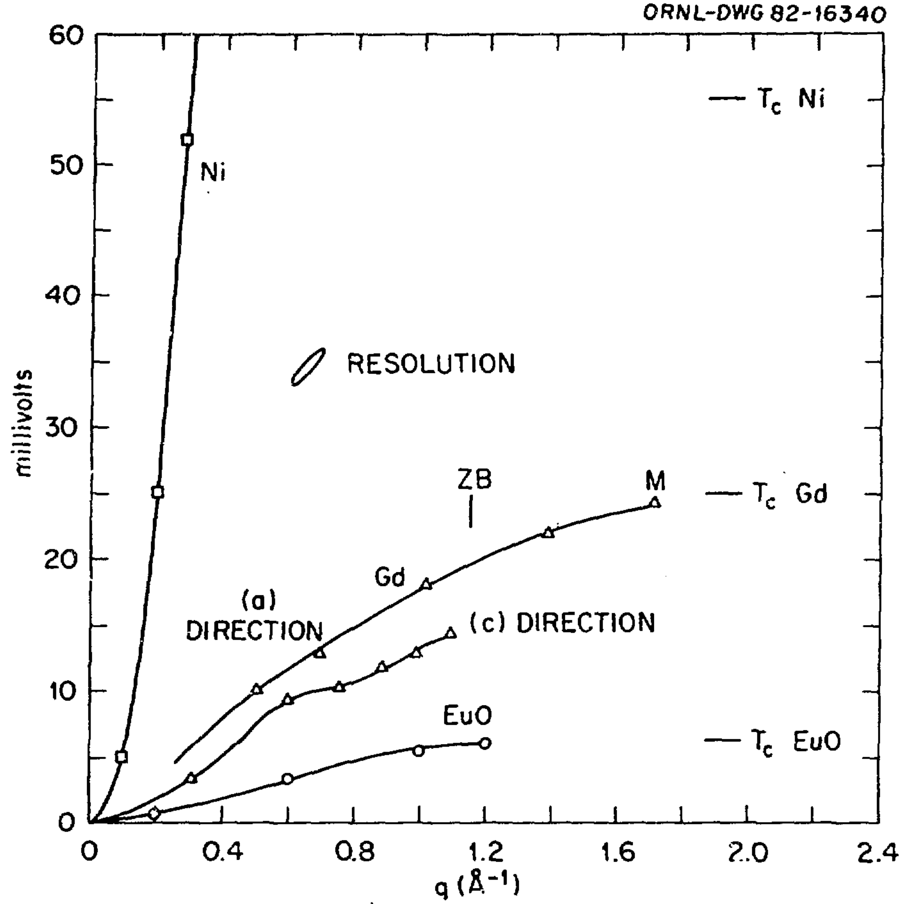


Fig. 4. Spin wave spectra for EuO, Gd, and Ni compared to their transition energies kT_c. Note that EuO and Gd become paramagnetic when all spin wave energies are excited but that kT_c for Ni is much lower than the spin wave energies.

then the isothermal susceptibility is given as

$$\chi_q \sim \frac{1}{K_1^2+q^2} \quad . \tag{24}$$

If further the time dependence of the spin correlations are diffusive in nature

$$\langle S^\alpha(q,0)\ S^\alpha(q,t)\rangle = \langle S^\alpha(q,0)\ S^\alpha(q,0)\rangle e^{-\Gamma q^2 t} \tag{25}$$

and

in this case

$$F(\bar{K}\omega) = \frac{1}{\pi} \frac{\Gamma}{\omega^2+\Gamma^2} \tag{26}$$

We then have that

$$S^\alpha(\bar{K},\omega) \sim \frac{1}{K_1^2+q^2} \frac{\Gamma}{\omega^2+\Gamma^2} \; . \tag{27}$$

Measurements above T_c for nickel have found a ridge of scattering that for higher energies looks similar to the scattering found below T_c. Figure 5 shows the temperature dependence of the center of this ridge. Uemura et al.[7] claim that the cross section derived from Eq. (27) explains this ridge for all measured energies. I disagree with this assertion as an equation based on diffusion results in line shapes unlike those observed in constant energy scans. Figure 6 shows measurements of spin waves in Ni for ΔE = 40 meV ~ 10 THz. I feel the dotted lines resulting from a diffusion based equation do not satisfactorily explain the observed data.

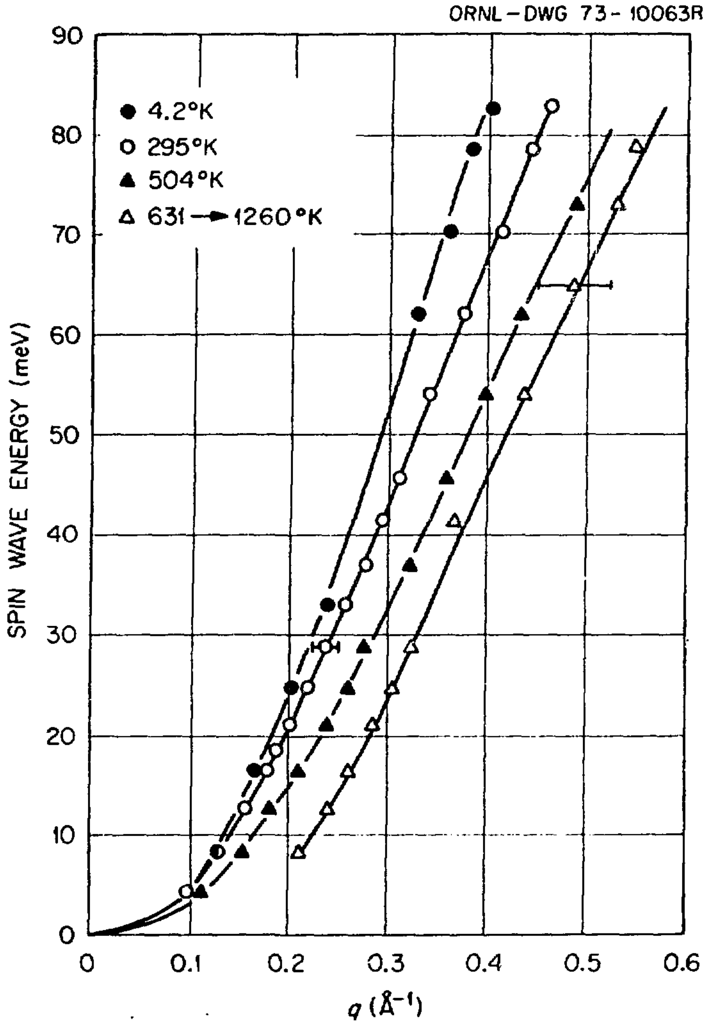
Because of the importance of neutron scattering from transition metals and the need for high energy neutrons for these studies, talks are to be given on this subject by Lynn and Uemura. No doubt differing viewpoints on the nature of the excitations in these materials above T_c will also be expressed.

The susceptibility of an isolated ion is given by

$$\chi_0 = 1/3\ (g\mu_B)^2\ S(S+1)\beta \; . \tag{28}$$

The isothermal susceptibility can then be obtained as below

$$\chi^\alpha_{\bar{K}} = \frac{\chi_0 \hbar}{N\ 1/3\ S(S+1)} \int_{-\infty}^{\infty} d\omega\ \frac{1 - e^{-\hbar\omega\beta}}{\hbar\omega\beta}\ S^\alpha(\bar{K},\omega) \; . \tag{29}$$



Ni Spin Wave Dispersion Relation.

Fig. 5. Magnetic excitation spectra for nickel at various temperatures. A ridge of scattering is found above T_c that for high energies is similar to the low temperature excitations.

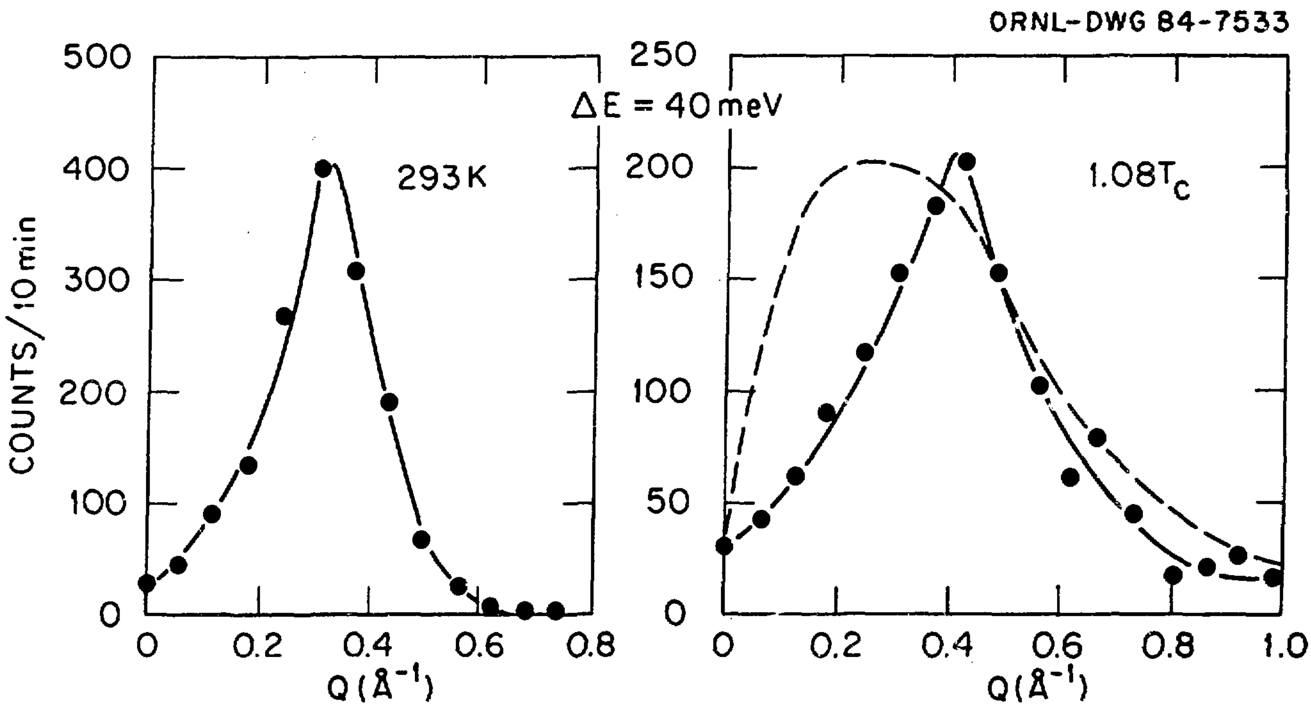


Fig. 6. Measurements of the magnetic excitations for nickel at room temperature and at 1.08 T_c. The measured line shape above T_c does not resemble that given by the diffusion equation as shown by the dashed line.

This gives a measure of the correlation range above T_c which is an important ingredient in choosing correct theories to describe the properties of transition metals. Capellmann will discuss experiments based on (29) but using polarized neturons to avoid nonmagntic processes.

Buyers will mostly discuss magnetic measurements in actinide systems. These systems have energies between rare earth and transition metals and incorporate the difficulties of both. The magnetic electrons can be rather itinerant in some materials and more localized in others. Certainly it is a rich area for the investigation of magnetic phenomena.

I hope that I have shown the power of neutron scattering techniques in investigating magnetism in a wide range of materials. The subject of magnetism is a difficult one and the joining of theory and experiments are gradually pointing directions toward solutions of these challenging problems. We hope that the new spallation sources will make possible additional measurements, particularly at higher energy transfers and thus add to our knowledge of magnetic materials.

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