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Temperature Dependence of the Magnetic Excitations in Nickel*

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The spin-wave excitations for nickel have been measured at a series of temperatures between 4.2 and 715°K. It is found that the spin waves change very little in character even at 80° above the Curie point. A sudden decrease in the spin-wave intensity at about 80 meV, interpreted as the intersection of the spin-wave spectra with a continuum band of Stoner excitations, is also found to be quite temperature independent.

Earlier neutron-scattering investigations of the spin-wave spectrum of nickel have shown that the spin-wave intensity falls off slowly with increasing energy until about 100 meV and then drops suddenly by an order of magnitude.[1] This sudden decrease in intensity was interpreted as being the result of the spin-wave excitation intersecting a continuum band of Stoner excitations. The intersection point was different for the three principal symmetry directions, the spin wave disappearing in the [111] direction first at about 80 meV and extending out the farthest in the [100] direction to about 110 meV. As the temperature of the sample is changed, it was expected that the splitting between the spin-up and spin-down bands would vary in a manner proportional to the magnetization and that the Stoner continuum would move to some degree as the band splitting was altered. In order to check this conjecture, we undertook a study of the spin-wave spectrum as a function of temperature. Since the high-energy spin waves are difficult to measure because of the low flux of high-energy neutrons from the reactor, it was decided to concentrate on the [111] direction where the spin waves disappear first. It was surprising for us to find that not only did the intersection point between spin waves and Stoner modes appear to be temperature independent, but that the whole spin-wave dispersion curve changed remarkably little between 4.2 and 715°K which is about 80° beyond the Curie point.

The magnetic excitations in nickel at various temperatures have been studied by Lowde and Windsor,[2] and their review article is a good source for reference to prior work in this field. Unfortunately, they were unable to employ enough resolution or neutron intensity to obtain a clear picture of the spin-wave excitations, especially at higher temperatures. They did observe some broad ridges in the neutron scattering at $1.1T_c$, which could be interpreted as collective excitations; however, they could say nothing about the high-energy spin waves that give information about the position of the Stoner modes. Minkiewicz *et al.*[3] made high-resolution measurements of the spin waves in nickel at temperatures up to the Curie point. They observed that for $q < 0.125$ Å^{-1} the spin waves became critically damped just below T_c and that no distinct spin-wave excitations could be observed at higher temperatures.

Our measurements were made on a triple-axis spectrometer installed at the high-flux isotope reactor. The sample used was a crystal of ^{60}Ni 1 in. in diameter by $1\frac{1}{2}$ in. long. The temperature was determined by means of two thermocouples welded to the sample. An additional check on the temperature was made by increasing the resolution of the spectrometer and measuring the critical scattering. An intensity-versus-temperature profile was obtained which is very similar to that shown by Minkiewicz *et al.*[3] in their Fig. 2, and this served to confirm the

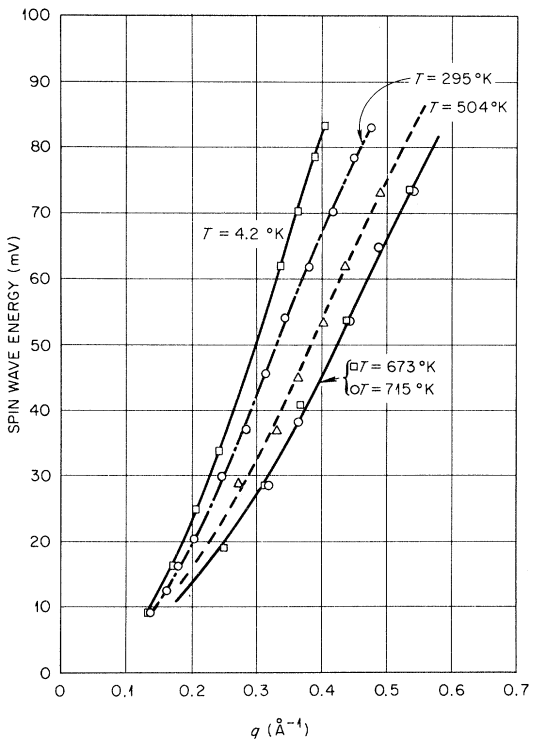

FIG. 1. Spin-wave dispersion in nickel as a function of temperature.

accuracy of the thermocouple readings. Some of our measured spin-wave spectra are shown in Fig. 1. Constant-energy scans were used because the steepness of the dispersion curves make constant-momentum-transfer scans difficult if not impossible. The measurements were made in a manner similar to those made on iron,[4] which have been described in detail, and thus the experimental technique will not be discussed at this time. We notice that the spin-wave spectra change very little from 4.2 to 715°K. The dispersion relation appears to renormalize with temperature up to T_c and then remain at a position independent of temperature. If the curves are approximated with a quadratic dispersion law $E = Dq^2$, one finds that D decreases from 555 meV Å^2 at 4.2°K to about 280 meV Å^2 at T_c and remains at this value as the temperature is increased. This is in clear contrast with the results of Minkiewicz *et al.*[3] for small q, which showed D decreasing to near zero as T approached T_c. Stringfellow,[5] using a small-angle scattering technique, found that D decreased to about 125 meV Å^2 at T_c; however, one would expect the triple-axis technique to be more reliable since the spin-wave excitations can be measured directly with no applied magnetic field.

We attempted to extend our measurements downwards to low enough energies so that our high-q data could be joined to the low-q data of Minkiewicz *et al.*[3] However, the interference of phonon branches prevented us from getting good data in this area. There is thus a small region in between the two sets of data that remains unexplored in any detail. Perhaps further measurements can be made in this region by using polarized beams to avoid the phonon scattering.

An example of some of the measured spin-wave peaks at elevated temperatures is shown in Fig. 2. The spectrometer resolution used in these measurements was fairly coarse in order to obtain a high enough counting rate to make accurate intensity measurements and little, if any, spin-wave broadening is observed as the temperature is increased. Higher-resolution measurements do show spin-wave broadening, however, and these measurements will be discussed at a later time. The spin waves are very well defined up to a temperature of 715°K which is as high in temperature as our furnace could go. Although spin waves appear to persist above the Néel temperature in some 3*d* antiferromagnetic materials and alloys,[6] it seems surprising that the spin waves are so distinct in a magnetically soft 3*d* transition metal like nickel.

The spin-wave intensities are more difficult to measure and interpret than the spin-wave positions. The neutron intensity obtained at the detector per monitor count is given by

$$i(\vec{Q}, E) = (c/\Delta V)\int S(\vec{Q}+\Delta\vec{Q}, E_{Q+\Delta Q})\,\delta(E + X_4 - E_{Q+\Delta Q})\exp(-\tfrac{1}{2}\textstyle\sum_{kl} M_{kl} X_k X_l)\,dX_1\,dX_2\,dX_3\,dX_4, \quad (1)$$

where $S(\vec{Q}, E)$ is the scattering law for the material, the X's are uncertainties in Q and E, and the M_{kl} are involved functions of the spectrometer parameters such as the d spacings of crystals and collimator divergences. The constant c involves further spectrometer parameters and angles and the efficiency of the monitor counter, and ΔV serves to normalize the resolution volume. The integrated intensity $I(E)$ of a neutron scan at energy E is obtained by integrating Eq. (1) over Q and to a good approximation is given by

$$I(E) \simeq cS(Q_0, E)/\eta = c\mu\beta F^2(Q_0) I_0(E)/\eta, \quad (2)$$

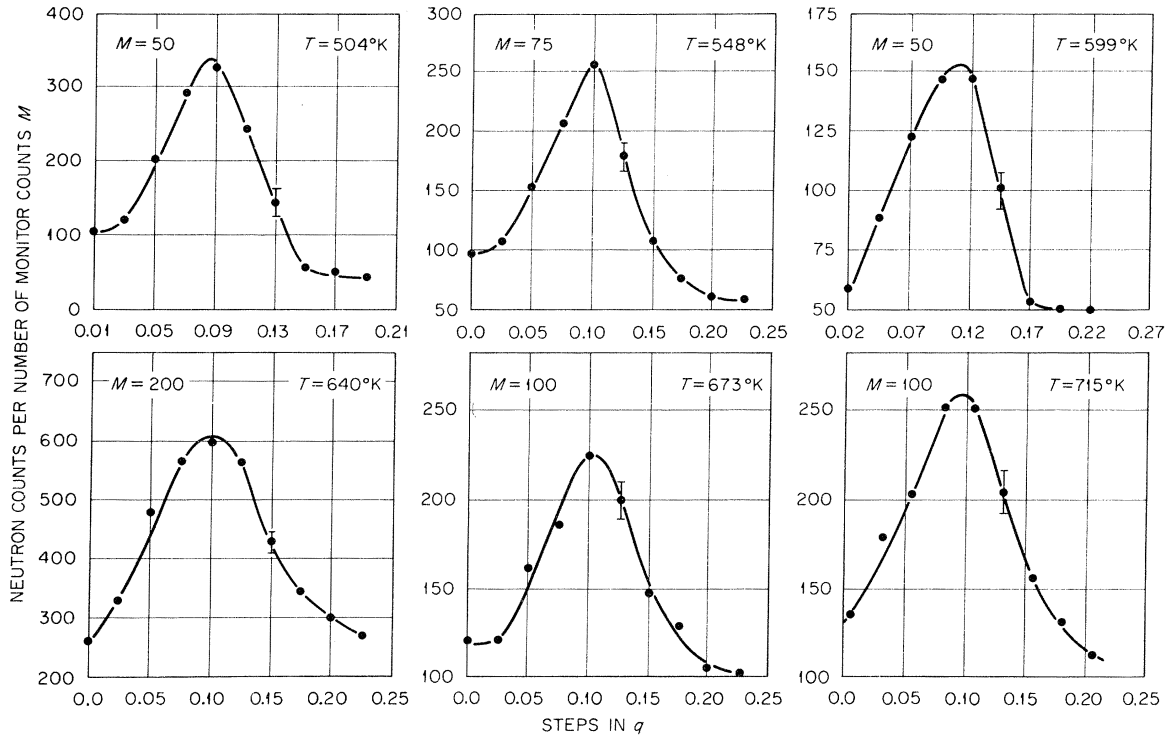

FIG. 2. Measured spin waves at a series of temperatures above and below the Curie point. The spin-wave energy is 29 meV and the measurements are in the [111] direction. q is plotted in zone-boundary units and must be multiplied by $2\pi\sqrt{3}/a \sim 3.09$ to obtain values in Å^{-1}.

where $F(Q_0)$ is the magnetic form factor, β is the Boltzman factor, μ is the moment on the nickel atom (assumed temperature independent), η is the slope of the dispersion curve, $I_0(E)$ is the spin-wave intensity, and Q_0 is the value of Q at the center of the scan. The integrated intensity $I(E)$ depends on the slope of the dispersion curve through the Jacobian factor discussed by Brockhouse *et al.*[7] and in Ref. 4. A detailed discussion of the method of obtaining integrated spin-wave intensities from constant energy-transfer scans performed on triple-axis spectrometers is given in Ref. 4, and thus the subject will not be discussed further at this time.

The spin-wave intensity $I_0(E)$ for various temperatures is shown in Fig. 3. It appears that the

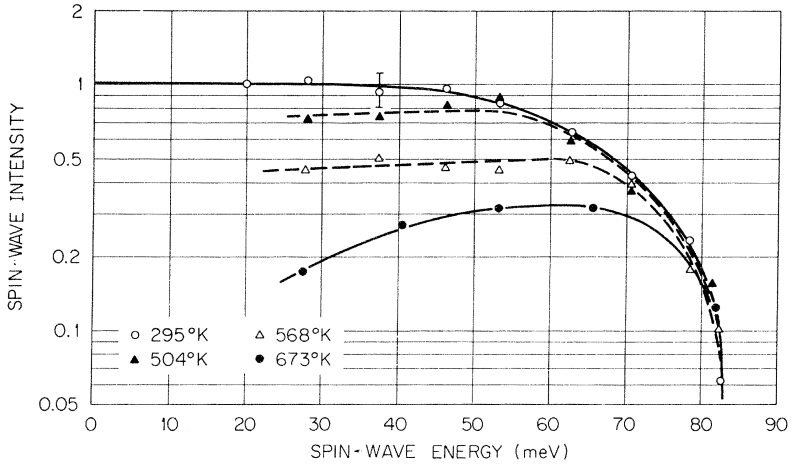

FIG. 3. Spin-wave intensity versus energy for the [111] direction in nickel at different temperatures.

sudden decrease in intensity interpreted as the intersection of the spin-wave and Stoner modes is temperature independent, although measurements at the higher temperatures are difficult since the spin-wave intensity is reduced at all energies. It is surprising that the intersection point does not decrease with temperature since one would expect the Stoner continuum to drop in energy as the band splitting decreases. We believe that our interpretation of the sudden decrease of spin-wave intensity as being due to Stoner modes is reasonable especially in view of the band calculations of Cooke[8,9] and Mook *et al.*[1] Certainly the falloff is intensity is not an artifact of the spectrometer since many measurements have been made to energies much higher than 90 meV in other materials, and in the [100] direction in nickel spin waves are easily measured past 100 meV. It is to be noted that even though the spin-wave dispersion curves are isotropic the Stoner continuum may be quite anisotropic. The band calculations clearly show this effect and in fact predict that the spin-wave intensity should fall off more rapidly in the [111] direction than in the [100] direction.[9] The existence of spin waves at high temperatures is perhaps reasonable if long-range spin correlations persist at elevated temperatures. It is not clear if the spin-wave Stoner band intersection can also be explained in these terms.

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Dipolar Interactions at Ferromagnetic Critical Points

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The critical behavior for $T > T_c$ and $H = 0$ of ferromagnets with both isotropic exchange coupling and dipolar interactions is studied by exact renormalization-group techniques in $d = 4 - \epsilon$ dimensions ($\epsilon > 0$) with ($n = d$)-component spins. A crossover from short-range isotropic (Heisenberg) to characteristic dipolar behavior occurs. The asymptotic spin-spin correlation function develops a factor $\delta_{\alpha\beta} - q^\alpha q^\beta/q^2$ which suppresses longitudinal fluctuations. Experiments on EuO and EuS are considered.

Magnetic dipole-dipole interactions exist in all magnetic materials but in most ferromagnets the ordering is caused by exchange coupling, which, in materials with $T_c > 300°\mathrm{K}$, strongly dominates the dipolar terms. We suppose the magnetic Hamiltonian can be written in the localized spin form

$$\mathcal{H} = -\tfrac{1}{2}\sum_{\vec{R},\vec{R}'}\left[J(\vec{R}-\vec{R}')\vec{S}_{\vec{R}}\cdot\vec{S}_{\vec{R}'} + (g_s\mu_B)^2\sum_{\alpha,\beta}\mathcal{A}^{\alpha\beta}(\vec{R}-\vec{R}')S_{\vec{R}}{}^{\alpha}S_{\vec{R}'}{}^{\beta}\right] \quad (1)$$

with, in d dimensions, the dipolar coupling

$$\mathcal{A}^{\alpha\beta}(\vec{R}) = d(R^\alpha R^\beta)/R^{d+2} - \delta_{\alpha\beta}/R^d, \quad (2)$$

while $J(\vec{R})$ is a short-range coupling of strength

$$\hat{J} = \textstyle\sum_{\vec{R}} J(\vec{R}). \quad (3)$$

Note that in (1) we take the number of spin components, n, equal to the dimensionality d, since the dipole-dipole interaction involves scalar products like $\vec{S}_{\vec{R}}\cdot(\vec{R}-\vec{R}')$ which couple spin and