

A droplet model for ferromagnetic spin waves above T_C

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Fisher's droplet model is used to calculate the neutron-scattering cross section for spin waves in a ferromagnet above T_C . Predictions are made for the wave-vector dependence of cross section, and a neutron-scattering experiment is proposed to test these predictions.

In order to explain the observation of spin waves and the apparent persistence of the Stoner splitting of spin-up and spin-down bands for temperatures above, as well as below, the Curie temperature in nickel¹ and iron,² a picture of short-wavelength magnetic excitations existing in clusters or droplets was invoked.³ Because the walls of these droplets are almost as thick as the droplets themselves for a spin rotationally invariant model (i.e., a low-anisotropy model such as the itinerant or Heisenberg model) the physical picture proposed was that of a slowly but continuously varying magnetization.^{3,4} On the basis of this physical picture, the polarized-spin neutron-scattering cross section for spin-wave creation was calculated, and a prediction was made for the magnetization dependence of the intensity.³ The picture of slowly varying magnetization was assumed to be valid as long as the average size of a droplet is large. Computer-simulation studies show⁵ that large droplets actually persist for temperatures well above the Curie temperature T_C . It is clear that simply assuming that the average size of a droplet is of the order of the correlation length is too naive because in reality we have a distribution of many different size droplets. Although at T_C the correlation length diverges, the size of all the droplets does not diverge. Rather, what diverges is the second moment of the droplet size distribution function (i.e., the susceptibility).

Fisher's droplet model⁶ should provide a qualitatively correct picture of the behavior of the distribution of such droplets as a function of temperature. Although this model may not be a quantitatively correct picture of critical phenomena, it is a simple model which may be a good starting point for describing magnetic excitations near and above T_C in ferromagnets.

Let us assume that independent excitations exist in each droplet and that the wave function of each excitation goes to zero at the wall of the droplet. Of course, this assumption is quantitatively incorrect (especially for an isotropic model in which the wall is

almost as thick as the droplet), but it is a simple boundary condition which will allow us to study qualitatively how requiring the excitations of a ferromagnet to obey boundary conditions at walls of each droplet affects the distribution of excitations as a function of wave vector. Naturally, this picture should be closer to the truth for anisotropic than isotropic magnets. Then, let us assume that the frequency and position-dependent magnetic susceptibility of a ferromagnet may be written in the droplet picture as

$$\chi(\bar{r}, \bar{r}', \omega) = \frac{1}{V} \int d^3R \int d\Omega \rho(\Omega) \times \chi_\Omega(\bar{r} - \bar{R}, \bar{r}' - \bar{R}, \omega) \quad (1)$$

where χ_Ω is the susceptibility at zero temperature of a region of volume Ω , $\rho(\Omega)$ is the number of droplets of volume Ω (the droplet distribution function), and \bar{R} is the location of the "center" of the droplet. Susceptibilities are defined so that the response in the magnetization to a field at point \bar{r}' is given by

$$\int d^3\bar{r}' \chi(\bar{r}, \bar{r}') H(\bar{r}') \quad .$$

We assume that χ_Ω vanishes unless \bar{r} and \bar{r}' are in the same droplet. What we are doing is averaging the susceptibility for a droplet of volume Ω and center located at \bar{R} over \bar{R} . The spin-wave contribution to χ_Ω (for a spin wave confined to a volume Ω with some boundary conditions on the walls of the droplet) can be written

$$\chi_\Omega(\bar{r}, \bar{r}', \omega) = \sum_{\bar{k}} \frac{\psi_{\bar{k}, \Omega}^-(\bar{r}) \psi_{\bar{k}, \Omega}^*(\bar{r}')}{\omega - \omega(\bar{k})} \quad , \quad (2)$$

$$\psi_{\bar{k}, \Omega}^-(\bar{r}) = \langle \bar{k} | S^-(\bar{r}) | 0 \rangle \quad , \quad (3)$$

and $\omega(\bar{k})$ is the spin-wave frequency. The spacing of allowed wave vectors \bar{k} is determined by the boundary conditions. The Fourier transform of χ defined in Eqs. (1) and (2) is given by

$$\chi(\bar{q}, \omega) = \frac{1}{V} \int d^3R \int d\Omega \rho(\Omega) \int d^3r e^{i\bar{q} \cdot (\bar{r} - \bar{r}')} \sum_{\bar{k}} \frac{\psi_{\bar{k}}^-(\bar{r} - \bar{R}) \psi_{\bar{k}}^*(\bar{r}' - \bar{R})}{\omega - \omega(\bar{k})} \quad . \quad (4)$$

This becomes

$$\chi(\bar{q}, \omega) = \int d\Omega \rho(\Omega) \sum_{\bar{k}} |g_{\Omega}(\bar{q}, \bar{k})|^2 \frac{1}{\omega - \omega(\bar{k})} \quad (5)$$

where

$$g_{\Omega}(\bar{q}, \bar{k}) = \frac{1}{\sqrt{V}} \int d^3r e^{-i\bar{q}\cdot\bar{r}} \psi_{\bar{k}}(\bar{r}) \quad (6)$$

For a finite droplet, the spin waves are standing waves and hence $\psi_{\bar{k}}(\bar{r})$ is a standing-wave function. For example, for a cubic droplet with the boundary condition that the wave function vanishes at the droplet wall,

$$\psi_{\bar{k}}(\bar{r}) = (1/\Omega) \sin k_x X \sin k_y Y \sin k_z Z \quad ,$$

where

$$\bar{k} = (\pi/\Omega^{1/3})(n_x, n_y, n_z) \quad .$$

The factor $|g_{\Omega}(\bar{q}, \bar{k})|^2$ is a function which is sharply peaked around $\bar{q} = \bar{k}$. Since its precise form depends on the shape of the droplet and all droplet shapes will occur, we will replace it by some convenient arbitrary peaked function. Although the width of this function depends on the droplet shape, the width for all globular droplet shapes should be the same order of magni-

tude. Then, let us consider a cubic droplet. From Eq. (6), we find that g is given by a product of three functions (one for k_x , one for k_y , and one for k_z) of the form

$$\frac{i}{2} \left[\frac{\sin(q_x - k_x)(\frac{1}{2}L)}{q_x - k_x} - \frac{\sin(q_x + k_x)(\frac{1}{2}L)}{q_x + k_x} \right] \quad (7)$$

where L is the length of a cubical droplet. This function is peaked about $\bar{k} = \bar{q}$ with a width of the order of $2\pi\sqrt{3}/\Omega^{1/3}$. Also, the minimum value of $|\bar{k}|$ allowed by the boundary conditions on a droplet of volume Ω depends on the droplet shape, but since it should have the same order of magnitude for all globular droplets, the value for a cubic droplet $\pi\sqrt{3}/\Omega^{1/3}$ will be used. Since the spacing of allowed values of \bar{k} depends on the droplet shape and since all possible droplet shapes occur, the distribution of \bar{k} values gets "smoothed out," and therefore, the sum over \bar{k} in Eq. (5) may be replaced by an integral. Also, since we do not know the specific form of g , let us replace it by an arbitrary function peaked at $k = q$ (for an infinite droplet, $|g|^2$ becomes a δ function). Then, let us use

$$\frac{\Omega}{(2\pi)^3} |g(\bar{q}, \bar{k})|^2 d^3k = \frac{\Omega\Gamma}{(q-k)^2 + \Gamma^2} \frac{dk}{4\pi q^2} \quad (8)$$

in Eq. (5). We find for the imaginary part of χ ,

$$\begin{aligned} \text{Im}\chi(q, \omega) &= \frac{1}{4\pi q^2} \int d\Omega \rho(\Omega) \frac{\Omega}{(2\pi)^3} \int d^3k |g_{\Omega}(q, \bar{k})|^2 \delta(\omega - \omega(\bar{k})) \\ &\quad \times \frac{1}{4\pi q^2} \int d\Omega \rho(\Omega) \Omega \int dk \frac{\Gamma}{\Gamma^2 + (q-k)^2} \delta(\omega - Dk^2) \quad . \end{aligned} \quad (9)$$

For relatively long-wavelength spin waves $\omega(k) = Dk^2$. The integral over k gives

$$\begin{aligned} \text{Im}\chi(q, \omega) &\sim \int d\Omega \rho(\Omega) \\ &\quad \times \frac{\Gamma}{\Gamma^2 + [q - (\omega/D)^{1/2}]^2} \frac{1}{4\pi q^2} \quad . \end{aligned} \quad (10)$$

Since the differential neutron scattering cross section is proportional to $q^2 \text{Im}\chi(q, \omega)$, the cross section is peaked around $q = (\omega/D)^{1/2}$ in this model, as it should be. For a cubic droplet, we estimate from Eq. (7) that

$$\Gamma = 2\pi\sqrt{3}/\Omega^{1/3}$$

and the minimum allowed value of k is given by

$$k_{\text{min}} = \pi\sqrt{3}/\Omega^{1/3} \quad .$$

Since for any shaped globular droplet k_{min} and Γ should be of the same form as for a cubic drop, we will use these values in Eq. (10). Therefore, in the integration over Ω there is a minimum value of Ω for each ω necessitated by the inequalities

$$(\omega/D)^{1/2} = k > k_{\text{min}} = \pi\sqrt{3}/\Omega^{1/3} \quad , \quad (11a)$$

$$\Omega > \pi^3 3^{3/2} (D/\omega)^{3/2} \quad . \quad (11b)$$

Let us use for the distribution in Ω the distribution function from Fisher's droplet model

$$\rho(\Omega) = \Omega^{-\tau} e^{-\Theta\Omega^\sigma} \quad , \quad (12a)$$

where

$$\Theta = |(T_c - T)/T_c| (1/a^{3\sigma}) \quad , \quad (12b)$$

where a is a lattice constant, $\tau \approx 2.3$ and $\sigma \approx 0.67$. Although Fisher's droplet model has its shortcomings, it contains what should be a qualitatively correct picture of the existence of a distribution of fluctuating clusters whose distribution function favors larger and larger clusters as T_c is approached. Converting the integral in Eq. (10) to an integral over Γ , we get

$$q^2 \text{Im}\chi(\bar{q}, \omega) \sim \int_0^{2q_0} d\Gamma \frac{\Gamma^{3\tau+2}}{\Gamma^2 + (q - q_0)^2} e^{(q_0/\Gamma)^{3\sigma}} \quad , \quad (13)$$

where $q_0 = (\omega/D)^{1/2}$ and $q_c = 2\pi\sqrt{3}\Theta^{r/3}$. In practice, because Eq. (12a) is only asymptotically correct for $\Omega \gg a^3$,⁶ Eq. (13) should only be valid for $1/a \gg q_0 > q_c$. A plot of the spectral function given by Eq. (13) is given in Fig. 1. The total intensity [the integral of Eq. (13) over q] is

$$\pi \int_0^{2q_0} d\Gamma \Gamma^{3\tau+1} e^{-(q_c/\Gamma)^{3\tau}} \quad (14)$$

A plot of intensity versus q_0/q_c is given in Fig. 2 for $q_0 > q_c$. The intensity as a function of q_0/q_c is approximately given by

$$\exp(q_0/q_c) \quad (15)$$

Thus, we see that the spectral density and hence the neutron scattering intensity becomes small as q_0 approaches q_c from above. On the other hand, it is easy to show on the basis of Fisher's droplet⁶ model that the reciprocal correlation length goes approximately as $\Theta^{(7-2)/2\sigma}$. Thus, we see that although the characteristic wavelength at which the spin-wave neutron scattering intensity drops off is different from the correlation length, both lengths approach infinity at $T = T_c$.

Although we do not expect this theory to be quantitatively correct, we expect the following qualitative results to be correct: There should exist highly damped spin waves above T_c of wavelength shorter than a characteristic length which approaches infinity as T_c is approached, with a temperature dependence different from that of the correlation length. As the wavelength of the spin waves approaches this characteristic length, the spin-wave scattering intensity should fall off quite rapidly. This paper also provides a physical picture of how the existence of a distribu-

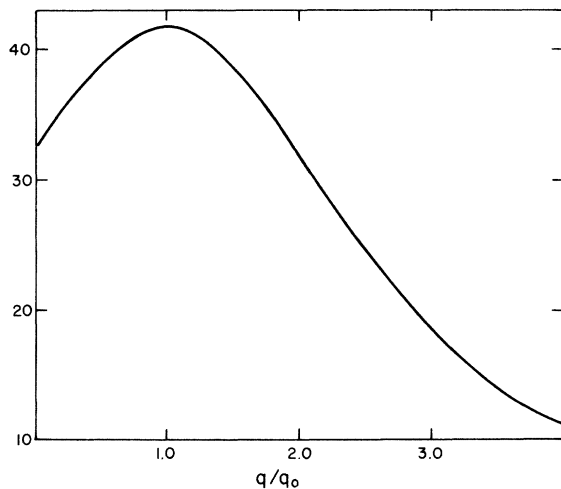


FIG. 1. Neutron scattering cross section (in arbitrary units) is plotted for $q_c/q_0 = 1$ as a function of q/q_0 .

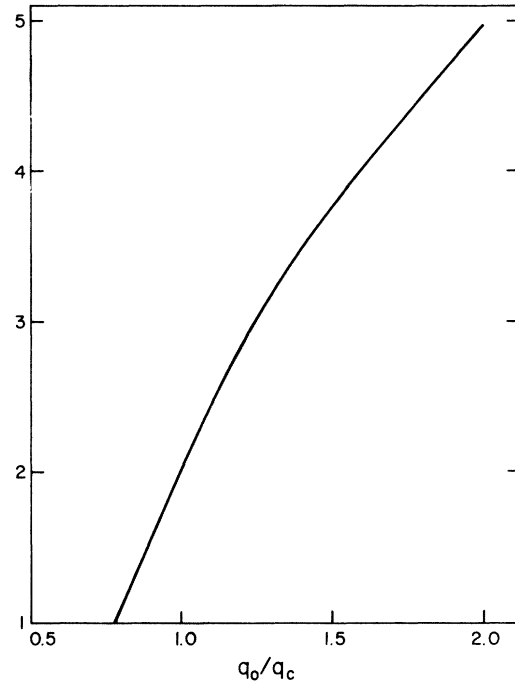


FIG. 2. Logarithm of the neutron scattering intensity (in arbitrary units) is plotted as a function of q_0/q_c .

tion of fluctuating clusters or domains above T_c affects the existence of and broadening of short-wavelength excitations.

Equation (9) has the form of the convolution of some broadening function with the spin-wave spectral density, very much similar to the broadening function introduced by Liu and Swanson and by Liu in their treatments of spin waves above T_c .⁷ In fact, if ω is set equal to zero in Liu's broadening function (i.e. the static limit taken), his results are qualitatively similar to ours in the sense that we both obtain a spectral function, which is a convolution of a broadening function with the zero-temperature spectral function, whose width increases as the spin-wave frequency increases. In this sense, this paper could be thought of as a type of microscopic justification of Liu's phenomenological theory.

The present theory, being a static one, should be valid as long as the spin-wave frequency is much greater than the decay rate of droplets of size greater than the spin wave q^{-1} . The decay time of a droplet should be larger than the time for a spin (pointed in a direction opposite to the magnetization of the droplet) to diffuse from the surface to the center of the droplet. On the basis of diffusion theory, this time is $(D^1/l^2)^{-1}$ where D^1 is the diffusion constant, which is of the order of the spin-wave stiffness D , and l is the droplet radius. From Eq. (11b), only droplets with

$l > q^{-1}$ contribute to the spin-wave neutron scattering. Since $D^1 q^2$ is the width of the central critical scattering peak, the criterion appears to be that the spin-wave frequency must be greater than the width of the central peak at wave vector q . Although the central peak width is not of the form $D^1 q^2$ for q^{-1} less than the correlation length according to dynamical scaling,⁸ the criterion for validity of the static theory that the spin-wave frequency be much greater than the central peak width should still be valid. In EuO and EuS (isotopic Heisenberg ferromagnets),⁹ the spin-wave frequency is always comparable to the central peak width, which is probably why spin waves are not seen in EuO above T_c as they are in Ni, for which the spin-wave frequen-

cies are much greater than the central peak widths for $q > 0.1 \text{ \AA}^{-1}$. (The spin waves fall within the central peak.)

Although the droplet model is not refined enough to predict in which materials spin waves will be seen above T_c , it can give the wave-vector dependence of the neutron-scattering cross section for materials in which they are seen.

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