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ABSOLUTE MEASUREMENT OF THE CRITICAL SCATTERING CROSS SECTION IN COBALT

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ABSTRACT

Small-angle neutron scattering techniques have been used to study the angular distribution of the critical scattering from cobalt above T_c. These measurements have been put on an absolute scale by calibrating the critical scattering directly against the nuclear incoherent scattering from cobalt. In this way the interaction range r_1, which appears in the classical and modified Ornstein-Zernike expressions for the asymptotic form of the spin pair correlation function and is related to the strength of the spin correlations, has been determined. We obtain $r_1/a = 0.46 \pm 0.03$ for the ratio of the interaction range to the nearest-neighbor distance in cobalt. This result is in good agreement with theoretical predictions. Lack of agreement among previous determinations of the ratio r_1/a made in iron failed to provide a definitive comparison with theory.

MASTER

INTRODUCTION

From the angular distribution of neutrons critically scattered from magnetic systems, considerable information can be obtained regarding the long range spin correlations which develop near the critical point. In the quasi-static limit[1] the scattered neutron intensity is directly proportional to the wave-vector-dependent susceptibility $\chi(q)$ which in the Orstein-Zernike approximation has the simple form

$$\chi(q)=\frac{\chi_o}{r_1^2(\kappa_1^2+q^2)}, \quad \chi_o=(g\mu_B)^2S(S+1)/3k_BT \qquad (1)$$

for small scattering vectors q. $\chi(q)$ is related to the spin pair correlation function $\gamma(r)$ by Fourier inversion, which, from Eq.(1), gives the well-known asymptotic dependence

$$\gamma(r)\propto\frac{1}{r_1^2}\;\frac{e^{-\kappa_1 r}}{r} \qquad (2)$$

for large distances r. The correlation range of the spin fluctuations, $\xi=\kappa_1^{-1}$, is obtained directly from the width of the Lorentzian shaped critical scattering and has been extensively investigated in numerous materials. However, to determine the parameter r_1, the so-called interaction range which is related to the strength of the spin correlations, measurements of $\chi(q)$ must be put on an absolute scale. For this reason there have been relatively few experimental determinations of r_1 to afford comparison with theoretical calculations.

In the course of detailed measurements of the small-angle scattering from polycrystalline, face-centered cubic cobalt, it was realized that cobalt is a particularly favorable system for an accurate measurement of the critical scattering cross section, and hence of the interaction range r_1. Absolute cross section measurements are usually carried out by normalizing the scattered intensity against that measured under identical experimental conditions from a sample whose cross section is well known. One difficulty with such a procedure, however, is that the scattering volumes of the two samples must be precisely known and taken into account when comparing intensities. Additional complications arise if the data must be corrected for multiple scattering which is often the case.[2] For cobalt, however, many of these difficulties may be circumvented by calibrating the critical scattering directly against the rather large and accurately measured[3] nuclear, spin-incoherent cross section (σ_{incoh} = 5.9 b) of cobalt itself. In this way factors of sample volume exactly cancel when forming intensity ratios. Furthermore, for the thin (2mm) slab sample geometry used in our experiments (necessitated by the moderately large absorption cross section of cobalt), analytic[2] and Monte Carlo calculations indicated that multiple scattering corrections were completely negligible for both the incoherent and critical scattering.

DESCRIPTION OF MEASUREMENT AND RESULTS

Our small-angle scattering measurements were made with a triple-axis neutron spectrometer using a mono-energetic incident beam of 13.5 meV. The angular dependence of the scattering was studied by operating the spectrometer in the double-axis mode in which all neutrons scattered through a fixed angle are detected, regardless of their energies. Energy analysis of the scattering was carried out in a separate series of triple-axis, constant-q scans. The wave vector and frequency dependence of the small-angle scattering in the vicinity of the critical point, obtained by correlating these two types of measurements, have been briefly described in a previous paper.[4]

Our absolute cross section measurement was predicated on being able to separate the intensity recorded in a double-axis scan into contributions due to magnetic scattering, nuclear incoherent scattering and that due to all other sources. This is partially accomplished quite simply by measuring the forward scattering at temperatures well below the critical point (T_c=1115°C), for example room temperature, where there is no magnetic scattering over the angular range of our double axis scans. At such temperatures the elastic magnetic scattering is concentrated entirely in Bragg peaks away from the forward direction. Inelastic magnetic scattering, i.e. spin wave scattering, also gives no contribution to the observed intensity as a fortuitous consequence of the extremely steep spin wave dispersion in cobalt. For a neutron to scatter from a spin wave through an angle θ, it must transfer energy $\hbar\omega$ given by the dispersion relation $\hbar\omega=\pm Dq^2(\omega,\theta)$ and also satisfy the usual momentum and energy conservation conditions. Near the forward direction the dispersion relation and conservation conditions can be satisfied simultaneously only for scattering angles $\theta<(\hbar^2/m_n)/D$. For cobalt at room temperature, the stiffness constant D∿500 meV-Å^2.[5] The spin wave scattering is therefore confined to angles θ<0.25° which lay outside the range of our angular scans.

The forward scattering which is observed at low temperatures is predominantly the elastic, nuclear, incoherent scattering. Inelastic nuclear (i.e. phonon) scattering is suppressed near the forward direction by the q^2 factor in the phonon cross section, and other possible sources of scattering, such as multiple Bragg and domain-wall scattering, are also expected to be small compared to the strong incoherent scattering. There is, of course, a contribution to the recorded intensity from sources other than the sample, such as scattering from the furnace which surrounded the sample, room background, detector noise, etc. To estimate this extrinsic background scattering, the following procedure was adopted. First, the transmission T of the cobalt sample was carefully measured to be 0.404 ($T = \exp(\rho\sigma_t\tau)$, where ρ is the number density, τ is the sample thickness, and σ_t is the total cross section which includes the absorption and incoherent cross sections, for example). Then knowing the incoherent cross section we could compute $T' = \exp[-\rho(\sigma_t - \sigma_{incoh})\tau] = 0.440$, which would be the transmission of the sample were there no incoherent scattering. A surrogate sample was then prepared by stacking foils of indium, having the same lateral dimensions as the cobalt sample, to a thickness which gave a measured transmission equal to T'. Indium was chosen for this purpose because it produces essentially no incoherent scattering but does have an appreciable absorption cross section. The room temperature, small-angle scattering observed from such an indium sample should then differ from that recorded for the cobalt sample only as a result of the incoherent scattering from the cobalt. By subtracting two such scans a a resultant uniform level of scattering was obtained which could then be associated entirely with the incoherent scattering.

Since the Debye-Waller factor in the neutron cross section is nearly unity in the forward direction, the small-angle incoherent scattering is effectively temperature independent. Hence the difference between identical angular scans taken at room temperature and at temperatures near T_c could be attributed entirely to magnetic critical scattering. Even very close to T_c, however, the critical scattering was observed to be inelastic to an appreciable degree. Because of this inelasticity, the total intensity measured at a fixed scattering angle represents a spread of scattering vectors q. Extensive triple-axis measurements of the energy distribution of the critical scattering at and above T_c did provide the necessary information to enable the effects of the inelasticity to be removed from our double-axis data in a manner which has been described in Ref. 4. When corrected for inelasticity, the angular dependence of our double-axis data is quite adequately described by the Ornstein-Zernike expression given in Eq.(1). This can be seen in Fig. 1 in which the reciprocal of the corrected double-axis intensity is plotted versus q^2 for several temperatures up to 100°C above T_c. The intercepts of the straight lines in Fig. 1 with the q^2-axis determine the value of κ_1^2 for each temperature.

After correcting for inelasticity, the critical scattering intensity measured in double-axis scans above T_c is directly proportional to the differential cross section,[6]

$$\left(\frac{d\sigma}{d\Omega}\right)_{crit} = \frac{2}{3}\, S(S+1)\left(\frac{\gamma e^2}{m_e c^2}\right)^2 |f(q)|^2 \, \frac{1}{r_1^2}\, \frac{1}{\kappa_1^2+q^2} \qquad (3)$$

where S is the effective spin of the scatterer, γ is the neutron magnetic moment in nuclear magnetons, and f(q) is the magnetic form factor. The same constant of proportionality relates the level of incoherent scattering deduced from the room temperature scans to the cross section

$$\left(\frac{d\sigma}{d\Omega}\right)_{incoh} = \frac{\sigma_{incoh}}{4\pi} \qquad (4)$$

Hence, ratios of the critical to incoherent intensities can be equated to those obtained by dividing Eqs.(3) and (4), evaluated at the corresponding wave vector q. Since the value of κ_1^2 in Eq.(3) is known from plots like those shown in Fig. 1 and $|f(q)|^2 = 1$ over the small-angle range of our data, these ratios yield absolute values for the interaction range r_1. Using for the value of the effective spin appropriate for face-centered cubic cobalt,[7] S=0.88, we obtain from our measured intensity ratios the result $r_1=1.16\pm 0.04\text{Å}$ for temperatures within 15°C of T_c. While r_1 varies from material to material, the ratio of r_1 to the nearest neighbor distance a_{nn} is expected to have universal significance. For cobalt near T_c, $a_{nn}=2.55\text{Å}$ so that

$$r_1/a_{nn} = 0.46\pm 0.03. \qquad (5)$$

Our value for the ratio r_1/a_{nn} is in good agreement with theoretical predictions, which have proven to be rather insensitive both to methods of calculation and to the crystal lattice. For example, the mean field approximation leads to the result[8] that $r_1/a_{nn} = 0.408$. High temperature series expansion calculations[9] based on the Heisenberg model give values for r_1/a_{nn} ranging from 0.45 to 0.52 depending on the spin and type of lattice; for a spin-1 system on a fcc lattice, which is the case most applicable to cobalt, $r_1/a_{nn} = 0.462$. Similar calculations[10] for the spin-1/2 Ising model yield values for r_1/a_{nn} from 0.44 to 0.46, the former number applying to an fcc lattice.

The few previous measurements of the ratio r_1/a are listed in Table 1. The lack of agreement among the results obtained for iron was a motivating factor for our measurement. The only previous determination of the interaction range in cobalt was made by Bally et al.[14] who normalized their neutron scattering data to bulk susceptibility measurements. Their result is in good agreement with our own, obtained by a completely different method. In view of our result, there now appears to be a discernable trend in the available data which indicates that experiment and theory are in basic accord.

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1. In the quasi-static limit the energy transferred by the scattered neutron is small compared to its incident energy.

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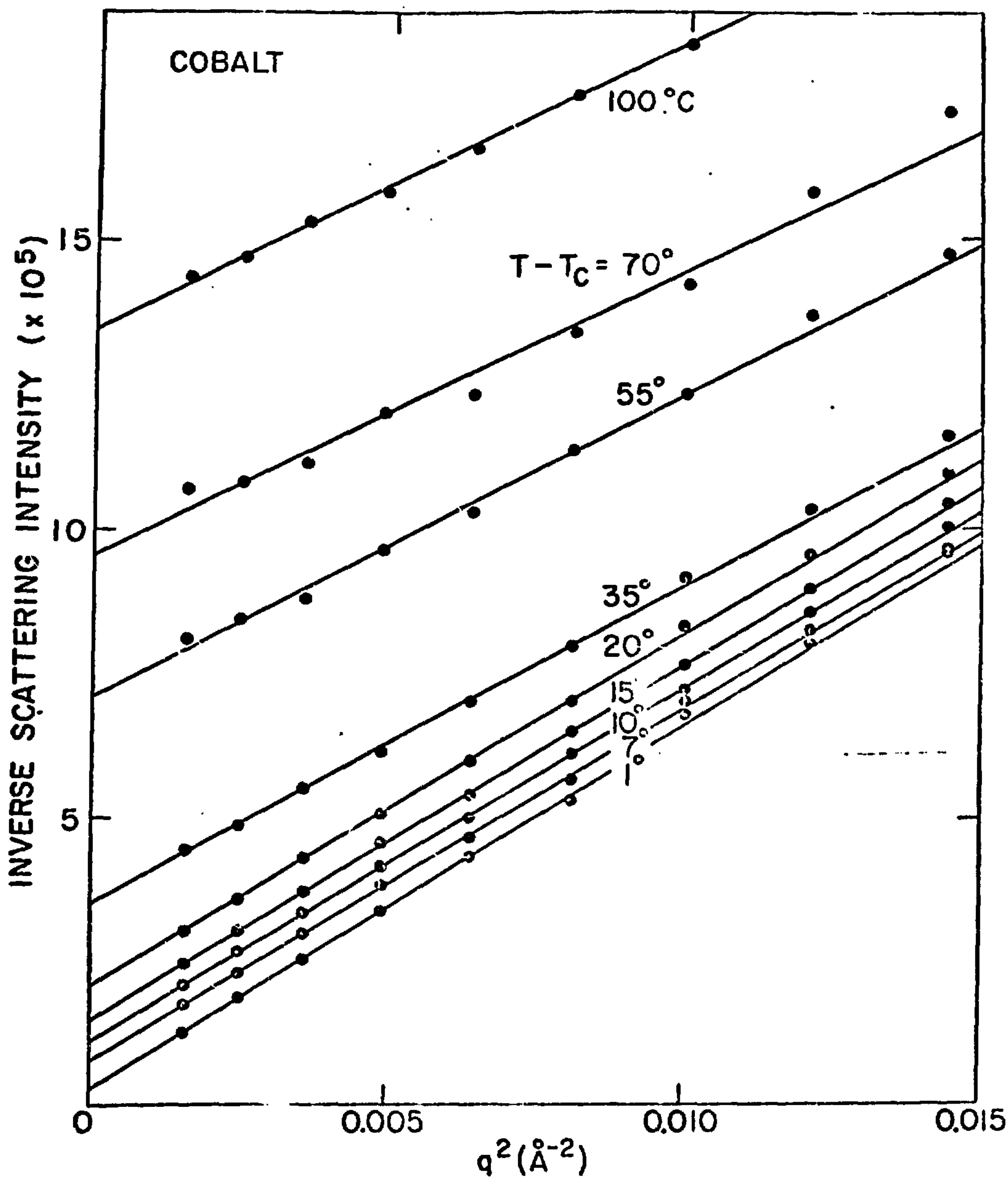
Table I. Results of previous measurements of the ratio of the interaction range r_1 to the nearest neighbor distance near T_c. $A=2.52$Å for iron and 2.55Å for cobalt.

r_1/a	Material	Reference
0.42±0.02	Iron	[11] Gersch, Shull, Wilkinson 1956
0.58±0.07	Iron	[12] Ericson and Jacrot, 1960
0.38	Iron	[13] Spooner and Averbach, 1966
0.43±0.04	Cobalt	[14] Bally et al., 1968

Fig. 1. Inverse intensities measured in angular scans above T_c plotted versus the square of the scattering vector after correcting for inelasticity.


COBALT
100 °C
T − T_C = 70°
55°
35°
20°
15
10°
7°
1°
INVERSE SCATTERING INTENSITY (x 10^5)
q^2 (Å^-2)
0
5
10
15
0.005
0.010
0.015