Polarized-Neutron Study of the Field-Induced Magnetic Moment in Chromium

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The polarization of chromium in a magnetic field is found to be of $3d$ electronic character. The data are best fitted by having a 60%-orbital–40%-spin contribution of the $3d$ electrons to the static magnetic susceptibility both above and below the antiferromagnetic transition temperature. The orbital contribution to the static magnetic susceptibility was found to be $(98\pm3)\times10^{-6}$ emu/mole. No evidence has been found of a well-defined localized spin above the antiferromagnetic transition temperature.

Since the discovery of antiferromagnetism in chromium by Shull and Wilkinson,[1] a considerable amount of experimental and theoretical effort has been directed towards an understanding of its properties. The electronic properties of chromium are of particular interest since it is generally accepted that they are associated with Overhauser spin-density waves.[2] In the present experiment, polarized-neutron techniques have been used to study the spatial distribution and temperature dependence of the magnetization induced in metallic chromium by an externally applied magnetic field. This investigation was undertaken in order to establish (a) whether the spatial distribution of the induced moment is appreciably different from that of a free ion, in view of the fact that chromium is considered to be a typical itinerant electron metal, (b) the electronic character of the field-induced magnetization in chromium and its distribution between orbital and electron spin polarization, and (c) whether the temperature dependence of the localized induced moment is characteristic of a well-defined intrinsic localized spin as has been suggested by NMR experiments.[3]

The quantity measured in a polarized-neutron experiment is the polarization ratio R, defined as the ratio of the coherent diffracted intensities for the two neutron spin states (parallel and antiparallel to the field on the crystal). The magnetic scattering amplitude $p(\theta)$ is simply related to the residual polarization ratio r:

$$r = R - 1 \simeq 4p(\theta)/b, \tag{1}$$

where θ is the Bragg angle and b is the coherent nuclear scattering amplitude (for chromium $b = 0.3532\times10^{-12}$ cm). The approximation is valid when $p(\theta) \ll b$, a condition fulfilled in our measurements ($r \sim 10^{-3}$–10^{-4} cm). In the present experiment the magnetic scattering amplitude consists of the paramagnetic,[4] diamagnetic,[5] and spin-orbit[6] scattering amplitudes. The correction to the measured polarization ratio introduced by the latter two quantities can be easily calculated and amounts to only 4% for the most severe case of the (110) reflection.

For intensity reasons the measurements have been performed with single crystals of high-purity chromium.[7] To minimize the secondary extinction correction the sample used in the measurements consisted of three 2-mm-thick slices. Each slice was deformed by compression until integrated reflectivity measurements indicated negligible secondary extinction for the composite-slice sample. The measurements, with 22 kOe of magnetic field on the sample, were performed in the 25–100°C temperature region using a highly polarized beam of 1.05-Å neutrons. Since the drifting in the diffraction peak intensity may introduce a sizable correction to the small residual polarization ratios under consideration, the neutron spin was flipped twenty times per second.

The angular dependence of the paramagnetic scattering amplitude, at 100°C, was obtained by measuring the polarization ratios for the (110), (200), (211), (220), and (310) reflections. The data, corrected for the contribution of the diamagnetic and spin-orbit scattering, are summarized in Fig. 1. It is seen that the induced-moment form factor decreases less rapidly with increasing angle than the magnetic form factor of the ordered state, determined from the intensities of antiferromagnetic reflections of Cr by Moon, Koehler, and Trego.[8] Thus, the induced-moment density distribution is less extended in space than the spin density in the ordered state. This suggests that a large part of the induced moment is of orbital origin. In order to see whether the induced-moment form factor could be interpreted in terms of *atomic* form factors, the experimental data were fitted using the free-atom $3d$ form factors of Freeman and Watson.[9] The

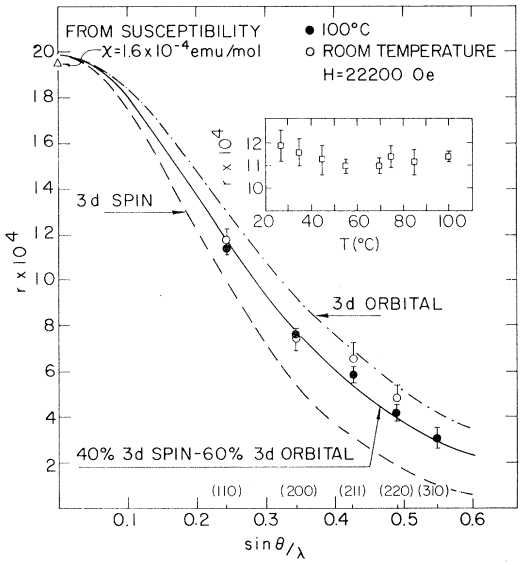

FIG. 1. Angular dependence of the induced-moment magnetic scattering amplitude compared with 3*d*-spin and 3*d*-orbital free-atom form factors. The 3*d*-spin curve is the magnetic form factor of chromium in the ordered state as determined by Moon, Koehler, and Trego (Ref. 8). The temperature dependence of the (110) magnetic scattering amplitude is shown in the inset.

data are best fitted by having a 60%-3*d*-orbital–40%-3*d*-spin contribution to the induced moment (Fig. 1). The zero-angle residual polarization ratio, obtained by extrapolation, is $(19.9 \pm 0.6) \times 10^{-4}$ and corresponds to a static susceptibility of $(163 \pm 5) \times 10^{-6}$ emu/mole. This value is to be compared with a total susceptibility of 160×10^{-6} emu/mole measured by the Faraday method on the same samples.[10] The close agreement between the total susceptibility and the 3*d* susceptibility obtained from the neutron-diffraction data implies that the 4*s*-electron contribution to the total susceptibility is small and comparable in magnitude to the chromium diamagnetism. The 3*d* spin and orbital susceptibilities, as determined from the analysis of the neutron diffraction data, are $(65 \pm 2) \times 10^{-6}$ emu/mole and $(98 \pm 3) \times 10^{-6}$ emu/mole, respectively. The large orbital contribution to the susceptibility implies that the gyromagnetic ratio g for chromium must be appreciably smaller than 2; in fact from our measurements $g = 1.25 \pm 0.04$. This is in excellent agreement with the value 1.21 ± 0.07 obtained[11] in a direct measurement, by the Einstein–de Haas method, of the gyromagnetic ratio of chromium. Preliminary measurements of the form factor at room temperature (Fig. 1) agree, to within experimental accuracy with those at 100°C. This implies that the induced moment does not change appreciably as the temperature is reduced below 40°C, the antiferromagnetic transition temperature of the sample.

The residual polarization ratios for the (110) Bragg reflection, in the 25–100°C temperature region, are plotted in the inset of Fig. 1. It is seen that the magnetic scattering amplitude is essentially temperature independent. Thus, to within experimental accuracy, we do not observe the conventional behavior expected for an intrinsic localized spin. This result is in disagreement with the interpretation, in terms of intrinsic localized moments, of NMR measurements[3] performed in the same temperature region on Cr^{53}. It should be mentioned that paramagnetic diffuse scattering experiments[12] at 210°C also failed to reveal the presence of any intrinsic localized spin in chromium. A possible explanation of the NMR measurements, which does not contradict the neutron diffraction results, is in terms of a spin fluctuation model.[13]

It is remarkable that the neutron-diffraction data in chromium, like in other 3*d* metals, may be interpreted in terms of free-atom form factors. The implication of this result may be easily understood if one notices that the induced-moment scattering amplitude is proportional to the off-diagonal components $\chi(\vec{Q}, \vec{Q}' = 0)$ of the generalized frequency-independent susceptibility function ($\vec{Q}$ is the neutron scattering vector). The spin contribution to the susceptibility function involves electronic states at the Fermi surface, whereas the orbital contribution is determined by matrix elements to excited electronic states. The neutron-diffraction data indicate that both contributions arise from 3*d* electronic states that can be approximated by tight-binding atomic orbitals. The orbital contribution of 98×10^{-6} emu/mole obtained from our data is in good agreement with band calculations of the induced orbital paramagnetism in chromium.[14] This is of considerable theoretical interest since the overall bandwidth enters rather directly in the calculation. The spin contribution of 65×10^{-6} emu/mole, on the other hand, is larger by a factor of ~3 than various calculated values of the unenhanced spin susceptibility.[15] This seems to indicate a surprisingly large exchange enhancement, but no definite conclusion may be drawn before more detailed band calculations of the spin susceptibility become available.

In conclusion, the angular distribution of the magnetic scattering in chromium can be described, as in other 3*d* metals, in terms of 3*d* free-atom form factors. The data are best fitted by having a 60%-3*d*-orbital–40%-3*d*-spin contribution to the induced moment both above and below the antiferromagnetic transition temperature. The orbital and spin susceptibilities, for our samples, were found to be $(98 \pm 3) \times 10^{-6}$ emu/mole and $(65 \pm 2) \times 10^{-6}$ emu/mole, respectively. These results are consistent with measurements of the total susceptibility and gyromagnetic ratio of chromium. The magnitude of the localized induced moment has been found to be essentially temperature independent, in the 25–100°C temperature region. Thus we do not observe the characteristic temperature dependence expected from an intrinsic localized spin.

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Neutron Scattering and the Correlation Functions of the Ising Model near T_c

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We study near T_c the correlation function $\langle \sigma_{00}\sigma_{MN}\rangle$ of the two-dimensional Ising model.

In this Letter we present the results of our computation of the Fourier transform of the spin-spin correlation function $\langle \sigma_{00}\sigma_{MN}\rangle$ for the two-dimensional Ising model in the scaling limit $T \to T_c$, $R = (N^2 + M^2)^{1/2} \to \infty$ with $(T - T_c)R$ fixed and of order 1.[1] We use these results to show that the asymptotic formula of Fisher and Langer[2] provides a much better method of extracting the critical exponent η from neutron scattering data than do the formulas of Fisher[3] and Fisher and Burford.[4]

More specifically, we have computed

$$F_{\pm}(t) = \lim R^{1/4}\langle \sigma_{00}\sigma_{MN}\rangle \tag{1}$$

in the scaling limit, where $t = \kappa R$, κ is the inverse correlation length [$\sim 2\ln(1+\sqrt{2})|T/T_c - 1|$ for the isotropic Ising model], and the subscripts +,− refer to above and below T_c, respectively. The momentum-dependent susceptibility is

$$\chi(\vec{k}, T) = \sum_{M=-\infty}^{\infty} \sum_{N=-\infty}^{\infty} e^{i\vec{k}\cdot\vec{R}}(\langle \sigma_{00}\sigma_{MN}\rangle - \mathfrak{M}^2), \tag{2}$$

where $\mathfrak{M}^2$ is the long-range order parameter. In the scaling region we define

$$X_{\pm}(y) = \lim_{k,\,\kappa \to 0} \kappa^{\gamma/\nu}\chi(\vec{k}, T), \tag{3}$$

where $y = k/\kappa$ is fixed and of order 1.

We have evaluated $X_{\pm}(y)$ and these are plotted in Fig. 1 for y not too large. For large y (> 20) the asymptotic expansions given below, in Eq.