

crystals.⁶ It was realized that the relatively simple mathematical formalism used in the nondirect model might just be an approximate substitute for the more complex formula appropriate for direct transitions.⁷ Later on, the photoemission of the noble metals was more often interpreted in terms of direct transitions. Most recently, Doniach⁸ proposed a many-electron theory of nondirect transitions. If applicable, the effect of this mechanism would be to reduce the intensity of the \bar{k} -conserving transitions. The more recent results on photoemission do not favor this interpretation.⁸ One important assumption which enters into the theory proposed by Doniach is a large effective mass of the electrons in the initial states. The results reported in this Letter refer to transitions between s - p bands with strong d admixture. However, the photon energy was not high enough to excite electrons from the flat d bands themselves. We hope that it will be possible to determine quantitatively the relative importance of nondirect and direct transitions from experimental results on the directional photoemission in the vacuum-uv region, where transitions from the d bands are important for photoemission of clean Cu single crystals.

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Critical Magnetic Neutron Scattering from CoO

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The critical magnetic neutron scattering from CoO has been measured near the Néel temperature T_N . The diffuse intensity at various wave vectors was assumed proportional to $A_+ |T/T_N - 1|^{-\gamma}$ above T_N and $A_- |T/T_N - 1|^{-\gamma'}$ below T_N . Using the method of Als-Nielsen, we measured the temperature dependence of the critical scattering at several values of q [the displacement from the (111) position in reciprocal space] and used these data to calculate the relative magnitudes of γ and γ' . In order to satisfy scaling law requirements we referred our measurements to a rigid lattice by correcting for the large tetragonal contraction below T_N . We found that $\gamma' = (1.02 \pm 0.02)\gamma$, and this suggests that static scaling laws may apply to CoO. In addition we ascertained that $A_+/A_- \cong 7.6 \pm 1.5$ in agreement with the linear theory of Schofield, where $A_+/A_- = 6.0 \pm 0.8$ for CoO.

We have recently presented magnetic neutron scattering measurements of the long-range antiferromagnetic order in CoO near the Néel tem-

perature.¹ The intensity of neutrons scattered by Co^{2+} ionic moments for the (111) magnetic peak varied as $I \propto (T_N - T)^{2\beta}$ in the critical region,

where $\beta = 0.244 \pm 0.015$. On correcting for the tetragonal lattice contraction below T_N , a value of $\beta = 0.29 \pm 0.025$ was obtained. This is within the range of the rigid lattice Ising prediction, $\beta = 0.3125$.

In this paper we report measurements on CoO of the diffuse magnetic neutron scattering (staggered susceptibility) as a function of temperature T in the critical region. We have measured the diffuse intensity in the vicinity of the (111) magnetic superlattice reflection at several positions q , where q is the difference between the superlattice position and the diffraction vector in reciprocal space. The intensity divergence in the critical region can be expressed in terms of simple power laws with critical exponents γ and γ' : $\chi(q=0, \epsilon_+) = A_+ \epsilon_+^{-\gamma}$ for $T > T_N$, and $\chi(q=0, \epsilon_-) = A_- \epsilon_-^{-\gamma'}$ for $T < T_N$, where $\epsilon_+ = |T/T_N - 1|$, $\epsilon_- = |T/T_N(T) - 1|$, and where A_+ and A_- are assumed constant on the basis of work by Schofield² and Schofield, Litster, and Ho.³ Fisher and Burford⁴ have indicated A_+ may not be constant but may vary with temperature. They suggest that A_+ might change as much as 6% over a reduced temperature range of $T/T_c = 1.0$ to 1.20. Very little is known of the critical scattering behavior below T_c , but if A_- also were a function of temperature, the combined effect of A_+ and A_- varying in opposite directions (if indeed they do) would affect our conclusions somewhat. The convention of plus and minus subscripts for the parameters refers to temperatures above and below T_N , respectively. The Néel temperature is expressed as a function of temperature below T_N in order to present our results in terms of the scaling-law formalism which assumes a constant volume. In order to accomplish this we express the Néel temperature as a function of temperature, $T_N(T)$, in the scaling law expressions. Using values of the pressure dependence of the Néel point⁵ and the compressibility, we may follow the approach of Heller⁶ and express the variation of T_N with temperature as

$$T_N(T) = T_N^0 \left[1 - 3.43 \frac{C(T) - C(T_N^0)}{C(T_N^0)} \right]. \quad (1)$$

The T_N^0 is the temperature at which the critical scattering is a maximum; $T_N^0 = 288.98 \pm 0.10^\circ\text{K}$ in this case, as determined by a least-squares fit. The $C(T)$ is defined as the tetragonal-axis length as a function of temperature and $C(T_N^0)$ the length at T_N^0 . Taking $\Delta C/C$ from strain measurements reported earlier, we corrected for the variation of T_N as a function of the tetragonal

lattice contraction below the Néel temperature.

The values of γ and γ' can be obtained from the temperature dependence of the critical scattering cross section above and below T_N , respectively, at the (111) position where $q = 0$. However, the measured intensity is a convolution of the true cross section and the instrumental resolution function; and it is necessary to unfold the measured intensity from the experimental resolution function to find $\chi(0, \epsilon_\pm)$. Since the resolution function lacked sufficient precision, we utilized the approach of Als-Nielsen⁷ to obtain relative values of γ and γ' at values of q for which the resolution function did not measurably affect the cross section. Moreover at these q positions there was a negligible Bragg-scattering component for $T < T_N$.

Neutrons of wavelength $\lambda = 1.05 \text{ \AA}$ were used, and a sharp collimation was utilized before and after diffraction; at the counter the horizontal divergence was 11.6 min and the vertical spread was about 35 min. The widths of the half-maximum intensity of the resolution function in the (h_1, h_1, h_3) plane of reciprocal space were 0.001 \AA^{-1} by 0.004 \AA^{-1} , and the width perpendicular to the plane was 0.011 \AA^{-1} . The crystal was nearly spherical with a mean diameter of 0.68 cm. The mosaic width was 7.1 min, and chemical analysis showed that the Co^{2+} -to- O^{2-} ratio was 1.001:1.000 with a 0.15% Ni^{2+} impurity.

Measurements of the critical scattering cross section were made at three positions on a circle through the (111) point in the (h_1, h_1, h_3) plane of reciprocal space at $q = 0.0071$, 0.0106 , and 0.0177 \AA^{-1} . Nuclear and magnetic-superlattice multiple-scattering contributions were negligible. The effect of the resolution function on the critical scattering intensity was determined to be less than 5%, except for some data taken at $q = 0.0071 \text{ \AA}^{-1}$. In the latter instance the cross section was unaffected for $\epsilon_+ > 0.03$ and $\epsilon_- > 0.007$. The intensities are shown in Fig. 1 for the three q locations as a function of the reduced temperature $[T/T_N(T) - 1]$. The uncertainty in the Néel temperature, $T_N^0 = (288.98 \pm 0.10)^\circ\text{K}$, introduces only a negligible error in the ratio $(\gamma - \gamma')/\gamma$ and is included in the estimate of the overall error.

Following the method developed by Als-Nielsen we expanded the critical scattering intensities about $q = 0$ in a power series to the second order in q . The dependence of the critical scattering on q above T_N is known,⁴ but the behavior below T_N is not well characterized. Als-Nielsen used power-series expansions in q to express the de-

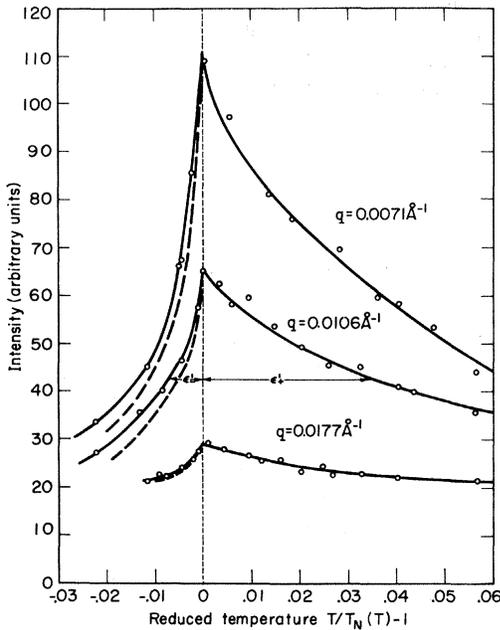


FIG. 1. Critical scattering intensities in the vicinity of (111). The solid lines are least-squares fits of the data by a power law representation of the intensities, corrected to a rigid lattice. The dashed lines are for the uncorrected data.

pendence of the diffuse scattering above and below T_N with different coefficients for each temperature region. In this approach the q dependence above and below T_N takes on a Lorentzian form since terms beyond q^2 are dropped. This is probably a reasonable assumption for CoO because experimental results^{1,8} have indicated this material appears to behave in an Ising fashion. For Ising systems it is expected that the critical scattering will be Lorentzian above and below T_N . We consider the parametric ratio between ϵ_+ and ϵ_-' , which are defined as the absolute values of the reduced temperatures above and below T_N , respectively, at which the cross sections are equivalent. This is illustrated in Fig. 1. The resultant relationship is

$$\frac{\epsilon_+}{\epsilon_-'} \cong \left(\frac{A_+}{A_-}\right)^{1/\gamma} \left[1 + \frac{\gamma - \gamma'}{\gamma \ln \epsilon_-'} - \frac{b_+}{\gamma} \left(\frac{q}{\kappa_+}\right)^2 \tau \right], \quad (2)$$

where b_+ is a coefficient of the q^2 term in the power series, κ_+ is the true inverse correlation range above T_N as given by Fisher and Burford,⁴ and $\tau = 1 - b_- \kappa_+^2 / b_+ \kappa_-^2$. We show ϵ_+ / ϵ_-' vs ϵ_-' for the three q positions in Fig. 2. We have included only those data points which are unaffected by the broadening function. Within the estimated uncertainty, the ratio ϵ_+ / ϵ_-' was indepen-

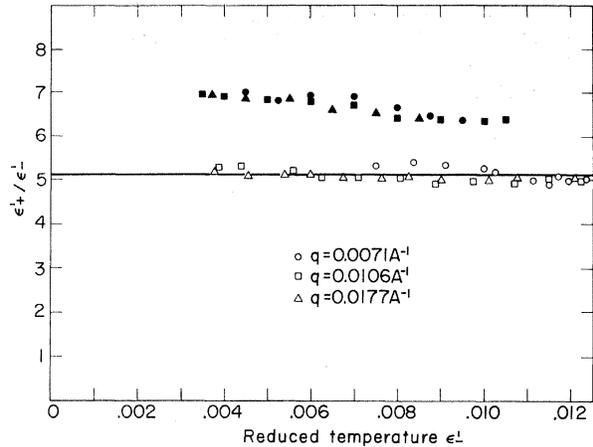


FIG. 2. Behavior of the ratio ϵ_+ / ϵ_-' . The open data points are corrected for the tetragonal distortion and the closed points are uncorrected.

dent of q , and this indicates that the last term in Eq. (2) is negligible. We also have shown ϵ_+ / ϵ_-' vs ϵ_-' in Fig. 2 for ϵ_-' values uncorrected for the nonrigid lattice. These data exhibit no q dependence either.

Assuming the Ising value of $\gamma = 1.25$, we used the data in Fig. 2 corrected for the nonrigid lattice to find $A_+ / A_- = 7.6 \pm 1.5$. The linear model of Schofield, Litster, and Ho³ gives $A_+ / A_- = (\gamma / \beta) [(1 - 2\beta)\gamma / 2\beta(\gamma - 1)]^{\gamma - 1}$. Utilizing our previously reported value $2\beta = 0.29 \pm 0.025$ and the assumed $\gamma = 1.25$, we found $A_+ / A_- = 6.0 \pm 0.8$, within range of our experimental ratio.

From the variation of ϵ_+ / ϵ_-' with ϵ_-' we found $(\gamma - \gamma') / \gamma = 0.02 \pm 0.02$, that is, $\gamma' = (1.02 \pm 0.02)\gamma$. Using the data uncorrected for the tetragonal deformation we obtained $\gamma = (1.17 \pm 0.02)\gamma'$. Our result corrected for the nonrigid lattice implies that the static scaling laws may be applicable to CoO since one condition of scaling is $\gamma = \gamma'$. However, numerical calculations⁹ have indicated that the symmetry of $\alpha = \alpha'$ and $\gamma = \gamma'$ may not hold for the three-dimensional models. The specific-heat exponents measured by Salamon⁸ for CoO are $\alpha = 0.12 \pm 0.01$ and $\alpha' = 0.05 \pm 0.02$, and this suggests that the scaling laws may not apply strictly.

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Tetragonal Elongation in CoO Near the Néel Point

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Measurements of the tetragonal elongation in a CoO single crystal in the critical region above the Néel temperature T_N indicate that $|\Delta L/L| \propto (T/T_N - 1)^x$, and below T_N that $|\Delta L/L| \propto (1 - T/T_N)^{x'}$, where $x = 0.67 \pm 0.10$ above T_N and $x' = 0.60 \pm 0.08$ below T_N . We compare these critical exponents with two theoretical predictions of strain behavior in the critical region and find agreement in both instances.

Above the Néel temperature ($T_N \cong 289^\circ\text{K}$) CoO is paramagnetic and has a NaCl structure. Below T_N CoO undergoes a phase transition from a paramagnetic to an antiferromagnetic state. This phase change is accompanied by a tetragonal contraction¹ with $c/a = 0.988$ at 93°K and a small shear deformation,² $e_{xy} = e_{yz} = e_{zx} = 5 \times 10^{-4}$. We present here measurements of the tetragonal contraction in the vicinity of T_N and characterize this contraction in terms of power laws: $|\Delta L/L| \propto (1 - T/T_N)^{x'}$ for $T < T_N$, and $|\Delta L/L| \propto (T/T_N - 1)^x$ when $T > T_N$. The critical exponents x and x' are compared, and the implications of our results are discussed in terms of the critical behavior of CoO.

Measurements of the strain were carried out along the [001] direction of a CoO single crystal using a dilatometer with a sensitivity of 10^{-6} cm. The crystal was cleaved along the {001} planes from a large boule grown by the flame fusion technique and annealed to relieve internal strains. Chemical analysis of the crystal gave a Co^{2+} -to- O^{2-} ratio of 1.001 to 1.000 with a 0.15% Ni^{2+} impurity. The cleaved crystal was mounted in the dilatometer such that the [001] crystal axis coincided with the dilatometer axis. The crystal was cooled through the Néel temperature by an ice-water bath following the method of Uchida *et al.*³ Most of the contraction was along the [001] direction, and this produced a nearly single-domain crystal. The temperature was controlled to $\pm 0.1^\circ\text{K}$, and 30 min were allowed for temperature stabilization of each point.

Only relative values are required to determine

the tetragonal elongation behavior in the critical region. There are only negligible normal elongations along the other two orthogonal axes as the tetragonal contraction occurs along the c axis.¹ In our measurements of the elongation we obtained approximately 92% of the tetragonal contraction along the [001] direction. Consequently, the small change in dimensions along the [001] direction for crystallographic domains with [100] and [010] c axis caused only a small absolute error. This error appears as a multiplicative constant in the power-law expressions and does not affect the determination of the critical exponents.

Figure 1 shows the tetragonal elongation for temperatures near T_N as well as data to 77°K . Above T_N there is a deviation from the elongation expected from the thermal dilation alone. This is associated with the presence of strong short-range magnetic correlation in the vicinity of T_N . Below T_N the tetragonal deformation is associated with the development of long-range antiferromagnetic order. The estimate of 92% single domain was made on the basis of the observed strain at 93°K . If we had a single domain, the elongation at 93°K would have been 0.0120.¹ In our case the observed contraction was 0.0106.

The data for $T < T_N$ were fitted by the power law $|\Delta L/L| = A(1 - T/T_N)^{x'} + B(1 - T/T_N)$ using the method of least squares; the second term arises from the thermal contribution. For $T > T_N$ we used the relationship $|\Delta L/L| = C(T/T_N - 1)^x + B(T/T_N - 1)$. We used T_N as a reference point and expressed $\Delta L/L$ in absolute values above and below