Critical Scattering in Cr + 0.2 at. % V and in Chromium

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The dynamic susceptibility $\chi(\mathbf{Q},\omega)$ of Cr + 0.2 at. % V near the Néel temperature, measured by neutron scattering, is found to be clearly incommensurate, in contrast to the apparently commensurate inelastic scattering seen in pure chromium. The paramagnetic state for both compositions is well described by a single form of susceptibility previously predicted from a two-band model of the itinerant-electron magnetism of chromium alloys, which naturally combines both commensurate and incommensurate diffuse scattering.

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The magnetic excitations and critical scattering near the Néel transition in chromium, as the archetypal itinerant-electron antiferromagnet,[1] have attracted considerable experimental[2-4] and theoretical[5-11] attention. In particular, the observation in pure chromium that the dynamic magnetic neutron scattering above the Néel temperature (T_N) is commensurate [centered on the (100) reciprocal-lattice position][4] while the ordered state that develops below T_N is incommensurate [magnetic Bragg peaks appear at $(1 \pm \delta,0,0)$ positions] has been difficult to understand.

Our understanding has also been hampered by the fact that the Néel transition that occurs in pure chromium is weakly first order.[12,13] However, alloying chromium with as little as 0.2% vanadium makes the transition continuous,[14,15] and this introduces the possibility of determining the critical behavior of a second-order transition into the incommensurate spin-density-wave (ISDW) state. A neutron-scattering experiment was therefore undertaken on a single crystal of each of Cr + 0.2 at. % V and Cr, to compare and contrast their dynamic susceptibilities near T_N. A key feature in understanding the results was the recognition of the relevance of expressions for the dynamic susceptibility of a two-band incommensurate itinerant-electron antiferromagnet presented by Sato and Maki.[9] We find that both samples in the paramagnetic state may be described by a relatively simple form proposed by Sato and Maki,[9] which corresponds to the sixfold symmetry around the nuclear reciprocal-lattice point.

The Cr + 0.2 at. % V crystal, of volume about 0.4 cm^3, was that used in an ultrasonic study of the Néel transition.[15] Properties of the Cr crystal, of volume about 0.2 cm^3, have been reported in several papers.[16-18] The experiments were performed on the C5 triple-axis crystal spectrometer at the NRU ractor at Chalk River, with Si(111) planes as both monochromator and analyzer, and a pyrolytic-graphite filter in the scattered beam of fixed wavelength 2.37 Å. Data were collected near the (100) reciprocal-lattice position in the (001) plane. Vertical slits of height 2.5 cm were mounted before the sample and the counter to limit the vertical divergence to 1.0°, and thus exclude scattering from the $(1,0,\pm\delta)$ ISDW Bragg peaks above and below the spectrometer plane. In the paramagnetic state, however, diffuse (i.e., critical) scattering centered on those positions will always contribute to scattering in the spectrometer plane to some extent, and must be included in the analysis of the data.

Elastic scattering as a function of temperature was used to locate the transitions. The *discontinuity* in intensity was clearly observed in Cr (see Fig. 1 of Ref. 19) at a nominal temperature of 310.1 ± 0.1 K, where the precision was limited by the apparatus temperature stability. No such discontinuity was seen in Cr + 0.2 at. % V, whose Néel temperature, nominally 288.7 ± 0.1 K, was identified as the point where the linewidth starts to increase above the resolution limit for a coherent Bragg peak (see Fig. 1 of Ref. 19).

To study the dynamic susceptibility, a set of nine temperatures for Cr + 0.2 at. % V, and five temperatures for Cr, was chosen in the range $T_N - 10$ K to $T_N + 20$ K. At each temperature, constant-ω scans at 0.0 (elastic), 0.5, and 1.0 THz and constant-$\mathbf{Q}$ scans (where $\hbar\omega$ and $\hbar\mathbf{Q}$ are the energy and momentum transfer) were taken through the (100) position and nearby satellite positions. Constant-ω data at 318 K in Cr are shown in Fig. 1, and at the corresponding temperature, i.e., $T_N + 8$ K, in Cr + 0.2 at. % V in Fig. 2. The constant-$\mathbf{Q}$ scans displayed Lorentzian energy dependence at all $\mathbf{Q}$ and $T > T_N$ that were chosen. The relative intensity from the two samples has been normalized by measuring and

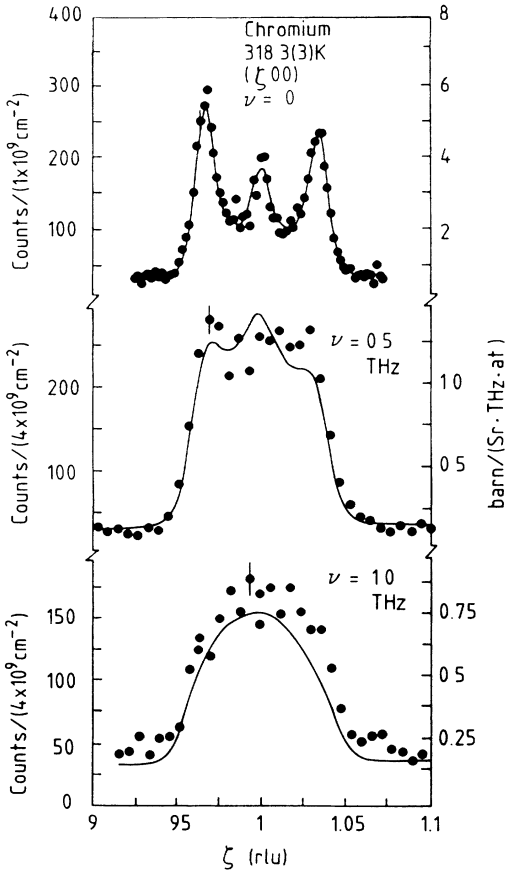

FIG. 1. Constant-energy triple-axis neutron-scattering scans of chromium at about 8 K above the Néel temperature. A typical counting error bar is shown on each scan. The left-hand scale is counts per indicated incident beam integrated over the time of one data point. The solid lines are the result of a fit of the Sato and Maki line shape described in the text simultaneously to all the data at this temperature, with the parameter values of the second row of Table I.

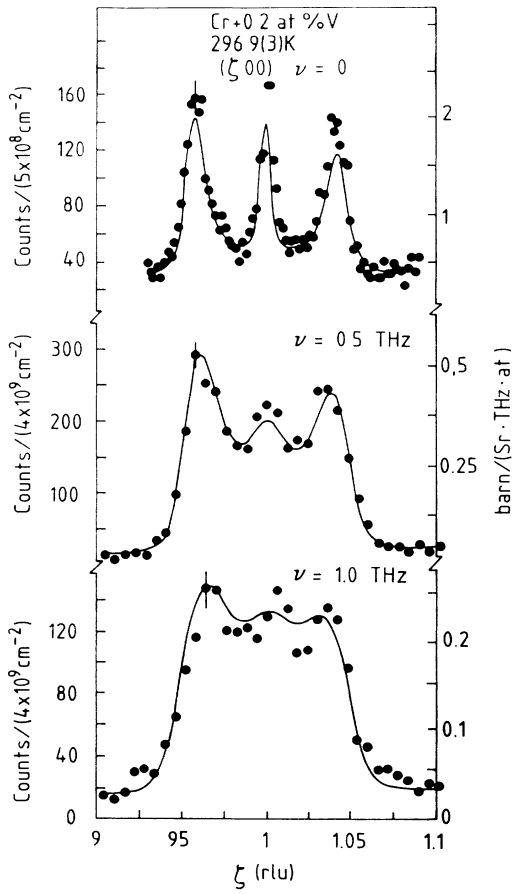

FIG. 2. Constant-energy triple-axis neutron-scattering scans of Cr+0.2 at.% V at about 8 K above the Néel temperature. The solid lines are the result of a fit of the Sato and Maki line shape described in the text simultaneously to all the data at this temperature, with the parameter values of the sixth row of Table I.

equating the phonon cross section, and incoherent elastic cross section, per atom (the two methods gave essentially the same normalization factor).

For critical scattering the imaginary part of the dynamic susceptibility[20] is written by Sato and Maki[9] as

$$\mathrm{Im}\chi(\mathbf{Q},\omega)=\frac{(\chi^0/r^2)A^2}{A^4[\kappa^2+R(\mathbf{q})]^2+\omega^2}, \qquad (1)$$

where χ^0 is the coefficient of the Curie-law susceptibility in the noninteracting limit, r is an interaction length scale, and A is the "magnetic stiffness." All three of these parameters are expected to be independent of temperature. κ is the inverse correlation length, which is temperature dependent, with a critical exponent ν that we observe to be consistent with $\frac{1}{2}$, the mean-field limit,[19] $\mathbf{q}$ is the offset from the ordering wave vector, and $R(\mathbf{q})$ is the real part of the self-energy. R is positive semidefinite, and is zero at the ordering wave vector. The generation of expressions like Eq. (1) by approximations to the general form of the susceptibility of an itinerant-electron system is discussed by Izuyama, Kim, and Kubo.[21] The simplest possible form for R is just $|\mathbf{q}|^2$, and this form has been used in the magnetic literature,[20,22] particularly for alloys of Mn in Cr in which the transition is second order to a commensurate antiferromagnetic state.[23] In the paramagnetic state of pure chromium six points near (100) correspond to the possible ordering wave vectors, and while our elastic-scan peaks center on those points, for energy transfers of 0.5 THz or more the scattering appears to be centered on the commensurate (100) position, as in previous work.[4]

In Cr+0.2 at.% V both the elastic and inelastic scans peak near the satellite positions, so we first attempted to fit the susceptibility with the superposition of the simple form described above at the six satellite positions near (100). This model, when folded with the spectrometer resolution function, does not fit the data well. The peaks of the observed scattering in constant-energy scans ap-

pear to be moving slightly toward the commensurate position as energy transfer increases, whereas this model line shape does not. The addition of a seventh, Lorentzian "commensurate diffuse," line does not significantly improve the fit.

Sato and Maki's[9] theory suggests the use of a single expression analogous to Eq. (1) with a self-energy function $R(\mathbf{q})$ that correctly represents the sixfold symmetry around the (100) position in the paramagnetic state. The simplest such function [their Eq. (35)] is a polynomial of fourth order in $\mathbf{q}$ involving only squares of the components of $\mathbf{q}$ with respect to the (100), (010), and (001) axes, where $\mathbf{q}$ is now the offset from the commensurate wave vector (100). The only such polynomial with continuous derivatives across (100) and zeros at the six satellite positions is

$$R(\mathbf{q})=\lambda_1[(|\mathbf{q}|^2-\delta^2)^2+\lambda_2(q_x^2q_z^2+q_y^2q_z^2+q_z^2q_x^2)], \quad (2)$$

where δ is the incommensuration below T_N, and λ_1 and λ_2 are parameters. The term in λ_2 provides the cubic anisotropy. If we assume R to be isotropic for small deviations from any one of the six minima, we obtain $\lambda_2=4$. The overall scale λ_1 can be absorbed in the other parameters in Eq. (1), and can be set to $1/4\delta^2$ to make $R(\Delta\mathbf{q})=|\Delta\mathbf{q}|^2$ for small deviation $\Delta\mathbf{q}$ from any one minimum. $A^2R(\mathbf{q})$ gives the excess energy of a fluctuation of wave vector $\mathbf{q}$ above that of a fluctuation at the incipient satellite wave vector. In particular, a fluctuation at $\mathbf{q}=0$ [$\mathbf{Q}=(100)$] has energy $A^2\delta^2/4$ above that minimum. The resulting expression for $\chi(\mathbf{Q},\omega)$ thus has an *intrinsic* response at the commensurate position, but its magnitude relative to that at the incipient satellite positions is related to the value of δ, a feature that does not arise for commensurate systems. When δ decreases, the susceptibility at the commensurate position increases relative to the peak at the incipient satellite position. Furthermore, for energy transfers less than $A^2\delta^2/4$ the susceptibility $\chi(\mathbf{Q},\omega)$ calculated from Eqs. (1) and (2) at the commensurate position is much smaller than that near the incipient satellite, and the scattering will be clearly incommensurate, while for $\omega\gg A^2\delta^2/4$, there is little difference in susceptibility between those two locations, and the scattering will be broadly commensurate.

Sato and Maki[9] stated the particular form above for Cr(Mn) compositions close to the commensurate-incommensurate triple point, and suggested that a more complicated form of $R(\mathbf{q})$ would be required for pure Cr. We find, however, that the dynamic susceptibility generated by Eqs. (1) and (2), folded with our spectrometer resolution, provides a remarkably good representation of the measured paramagnetic response, both elastic and inelastic, for both Cr+0.2 at.% V and Cr. Each of the rows of Table I shows the result of a simultaneous fit of the model to all of the data at that temperature, constant ω (elastic and inelastic), and constant $\mathbf{Q}$, an average of 165 data points per fit, with an average χ^2 of 1.9 per degree of freedom. Each figure shows part of the data at the temperature indicated, and the part of the corresponding fit as solid lines. The model thus provides the link previously missing between the static and dynamic correlations in paramagnetic Cr.[4] In the language of this model, the neutron-scattering data in the paramagnetic state of Cr have been difficult to interpret because the commensurate fluctuations are higher in energy than the fluctuations at the critical wave vector by only $A^2\delta^2/4=1.3$ THz. This is a small difference since the Néel transition temperature corresponds to an energy of 6.5 THz. In Cr+0.2 at.% V, δ is larger, and the commensurate energy difference is thus increased, to $A^2\delta^2/4=2.1$ THz, so the commensurate response is correspondingly reduced. The different qualitative form of the inelastic scattering is indirectly the result of the increase in the incommensurability in the vanadium-doped sample.

The results summarized in Table I are at too few temperatures to estimate any critical exponent for Cr+0.2

TABLE I. Parameters of Eqs. (1) and (2) deduced from fits to triple-axis neutron-scattering data above T_N. χ^0/r^2 is the amplitude scale. Numbers in parentheses are statistical uncertainties in the final digits.

Temperature (K)	$10^3\chi^0/r^2$ (μ_B^2/Å^{2}at.)	A^2 (THzÅ^2)	δ (Å^{-1})	κ^2 (10^{-5} Å^{-2})
		Chromium		
312.4(4)	1.13(4)	840(60)	0.0803(3)	2(1)
318.3(3)	1.21(4)	800(60)	0.0780(5)	9(2)
328.5(3)	1.37(6)	770(60)	0.0773(6)	25(3)
		Cr+0.2 at.% V		
290.0(2)	0.53(2)	1030(50)	0.0947(2)	0.6(6)
291.8(2)	0.57(2)	980(40)	0.0947(3)	3.2(9)
296.9(3)	0.59(2)	960(50)	0.0944(5)	12(1)
297.8(1)	0.60(2)	930(50)	0.0939(5)	14(2)
308.0(1)	0.68(3)	870(50)	0.0930(8)	36(4)

at.% V reliably. An analysis in terms of critical effects must include our additional elastic-scattering data near the Néel temperature, and our inelastic-scattering data at temperatures below T_N.[19] Furthermore, the fit values of χ^0/r^2, A^2, and δ are monotonic functions of temperature in Table I, an additional complication. Since the ordering wave vector is a function of temperature below T_N, it is not surprising that δ, its extrapolation into the paramagnetic state, is also temperature dependent, but usually critical-effects analyses assume that the ordering wave vector is a fixed point in reciprocal space. Explanation of the temperature dependence of the wave vector below T_N requires detailed consideration of the incommensurate magnetic interaction in the chromium bands,[24] and a similar theoretical study of the other parameters in Table I may predict temperature dependence for them as well. Thus the complete analysis of the neutron-scattering data may require comparison to calculations involving the particulars of the itinerant-electron magnetism in chromium (which was the spirit of Ref. 9).

We have shown that the paramagnetic critical fluctuations in Cr+0.2 at.% V are similar to those in Cr, even though the order of the Néel transition is different in the two. The paramagnetic susceptibility of both materials can be understood in terms of the two-band itinerant-electron model of the magnetism of chromium, by use of the true symmetry of the paramagnetic state. The commensurate paramagnetic-fluctuation "problem" in chromium is found to be due to a quite small energy difference between commensurate and incommensurate fluctuations in the framework of Sato and Maki's theory.

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