Observation of Incommensurate Spin-Density-Wave Paramagnons

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Magnetic fluctuations seen by inelastic neutron scattering in the paramagnet $Cr_{0.95}V_{0.05}$ have a structure factor similar to that of the static incommensurate spin-density wave (SDW) in pure Cr, and are thus termed SDW paramagnons. Commensurate-diffuse scattering dominant in the paramagnetic phase of Cr is absent in $Cr_{0.95}V_{0.05}$. The amplitude of the SDW paramagnons in this alloy increases with temperature and their energy scale extends beyond 40 meV.

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We have observed for the first time, by inelastic neutron scattering, magnetic fluctuations in a paramagnetic transition metal alloy ($Cr_{0.95}V_{0.05}$) which peak at a wave vector incommensurable with the reciprocal lattice. This fluctuating spin-density wave[1] is termed a spin-density-wave (SDW) paramagnon.[2] The dynamic structure factor $S(\mathbf{q},\omega)$ corresponding to the SDW paramagnons is observed to increase with temperature, by a factor of about 2 at 300 K, relative to the low-temperature value. The magnetic scattering cross section is very small, being a factor of at least 100 less than in pure Cr.

The $Cr_{0.95}V_{0.05}$ single crystal of volume about 10 cm^3 (diameter of $\simeq$1.5 cm) was cut from a boule supplied by F. A. Schmidt, Materials Research Center, Iowa State University. It was prepared by arc-zone melting. The mosaic width is 1.2°. The neutron-scattering experiment was performed on the triple-axis spectrometer H7 at the Brookhaven high-flux beam reactor. A pyrolytic-graphite (002) monochromator and analyzer was used, with collimation 40′-40′-80′-80′ according to the usual convention. The data reported in this paper were taken with a fixed final neutron energy, E_f=14.7 meV. The crystal was mounted for scattering in the $(hk0)$ reciprocal-lattice plane in a Displex refrigerator.

The addition of small amounts of vanadium to bcc chromium rapidly depresses the Néel temperature (T_N =312 K) and the rms ordered moment, and at about 4 at.% V, T_N=0 K.[3] The wave vector, **Q**, of the spin-density wave moves further from the commensurate [100] point as V is added. In pure Cr, $Q=0.951a^*$ (at T=0 K), and at 2.5 at.% V it is $0.922a^*$, where $a^*=2\pi/a$; the variation is slower than linear (see Fig. 6 of Koehler *et al.*[4]). Elastic scans along the [100] axis in our $Cr_{0.95}V_{0.05}$ crystal at 9 K show no peaks at the satellite positions $[1\pm\delta,0,0]$. However, for inelastic constant-ΔE scans in this region of reciprocal space, well-defined peaks are seen in both longitudinal (L) and transverse (T) scans [see Fig. 1(a)] at the incommensurate points, as shown in Figs. 1(c) and 1(d). Aside from the much smaller magnitude of the scattered intensity here and the precise location of the peaks, these data look very similar to the constant-ΔE scans in pure Cr at low temperatures.[6] In pure Cr these peaks in the inelastic scattering are attributed to spin waves, which can be viewed classically as precessional modes of the 3*d* atomic moments, but with amplitude decreasing rapidly with increasing excitation energy $\hbar\omega$. By contrast, in $Cr_{0.95}V_{0.05}$, satellites are not seen in the elastic scattering (otherwise, the alloy would not correctly be described as paramagnetic), and the magnitude of the scattering increases with increasing energy transfer, as shown in Fig. 1(b). Fitting the data in Fig. 1(d) by two Gaussian peaks gives a value for the incommensurability parameter of $\delta=0.084\pm0.002$, so that the SDW wave vector is $Q=(0.916\pm0.002)a^*$. This value is consistent with the variation of Q with vanadium concentration obtained from the SDW Bragg peaks in the lower-concentration long-range-ordered alloys.[4] To within experimental accuracy ($\pm0.005a^*$), the widths of the peaks in Fig. 1 do not change with increasing excitation energy. The data require an excitation spectrum having a slope (ω vs q) of greater than 1 eV Å.

We have followed the temperature dependence of this paramagnetic scattering up to room temperature. Longitudinal and transverse scans at 299 K are shown in Fig. 2. It is seen that the magnetic scattering cross section increases by a factor of about 2. Fitting each of these scans by two Gaussian peaks, we find that the incommensurability parameter decreases to a value δ =0.066 ± 0.002, so that at room temperature $Q=(0.934\pm0.002)a^*$. Thus Q increases by about (1.4 ± 0.2)%, nearly identical to the variation of the wave vector of the static SDW in pure Cr between 0 and 300 K. Even at 299 K the magnetic scattering cross section increases with excitation energy $\hbar\omega$ as shown in the inset to Fig. 2. Fixing the final energy of the spectrometer at 30.5 meV, we have attempted to measure the spectral distribution of the magnetic intensity up to $\hbar\omega$=42 meV at several temperatures. This proved to be difficult because of the incoherent phonon scattering and the occurrence

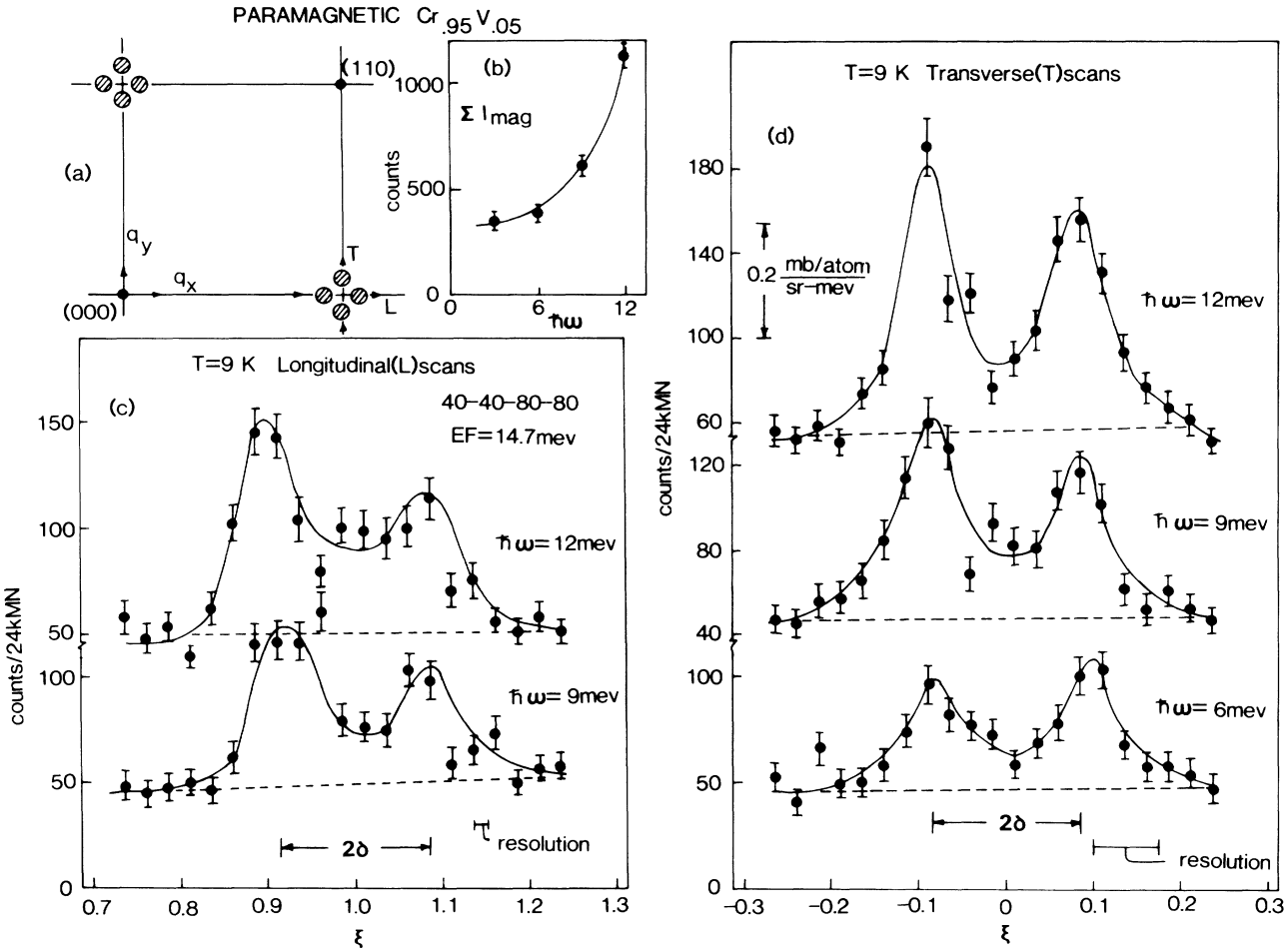

FIG. 1. (a) Reciprocal-lattice diagram showing the highly localized regions of **q** space, centered on incommensurate points, where paramagnetic scattering is observed in $Cr_{0.95}V_{0.05}$. (b) The integrated magnetic scattering, called $\sum I_{mag}$ here, vs excitation energy $\hbar\omega$, corresponding to the data in (c) and (d) at temperature $T=9$ K. (c) Longitudinal constant-ΔE scans at 9 K. The scattering vector is $\mathbf{q}=[\xi,0,0]a^*$. The peak at $\xi=0.916$ is more intense than the peak at $\xi=1.084$, because the falloff of the Cr $3d$ magnetic form factor $f(\mathbf{q})$ with **q**. (d) Transverse constant-ΔE scans at 9 K. The scattering vector is $\mathbf{q}=[1,\xi,0]a^*$. The counting time per point is about 8 min (kMN means thousands of monitor counts). The spectrometer energy resolution is 1.4 meV. The **q** resolution is 0.02 $Å^{-1}$ for longitudinal scans, and 0.14 $Å^{-1}$ for transverse scans. The resolution perpendicular to the scattering plane is 0.105 $Å^{-1}$. Here $a^*=2\pi/a=2.17$ $Å^{-1}$. The absolute calibration of the scattered intensity (in mb/sr·meV·atom) is obtained by normalization to a transverse-acoustic-phonon measured at $\mathbf{q}=[1.1,0.9,0]a^*$. The resolution volume correction (Appendix of Ref. 5) was not made here. The large background is due to the sizable incoherent scattering cross section (2.04 b/atom).

of the zone-boundary LA phonon at 30 meV at (100). It is clear from our limited data, however, that the magnetic scattering has a characteristic energy scale (width Γ) greater than 40 meV at 150 K.

It is truly remarkable that the incommensurate SDW instability at $2k_F$ in the electron gas of chromium metal manifests itself so clearly in these neutron-scattering measurements of the dynamic susceptibility $\chi(\mathbf{q},\omega)$ of this paramagnetic Cr alloy all the way up to room temperature. This observation is even more striking when contrasted to neutron-scattering measurements on pure Cr. Previous studies[5-7] of the paramagnetic state of pure Cr, up to temperatures about twice the Néel temperature T_N have shown that the neutron scattering is centered at [100], and is confined to a small spherical region about this commensurate point, having a diameter less than $\sim 0.07a^*$ as shown in Fig. 6 of Ref. 5. This so-called commensurate-diffuse (CD) scattering first appears more than 100 K below the Néel point. It has been suggested that it is due to phase fluctuations of the static SDW,[5,8] which ultimately lead to the disappearance of the long-range-ordered state at the Néel point. The Lorentzian spectral width parameter Γ increases rapidly from 3.2 meV at T_N to 15.6 meV at 500 K (see Figs. 14 and 15 of Ref. 5). Thus, the character of the paramagnetic scattering in pure Cr is really quite different from that in $Cr_{0.95}V_{0.05}$; its magnitude is more than a factor of 100 greater, it is centered at the commensurate

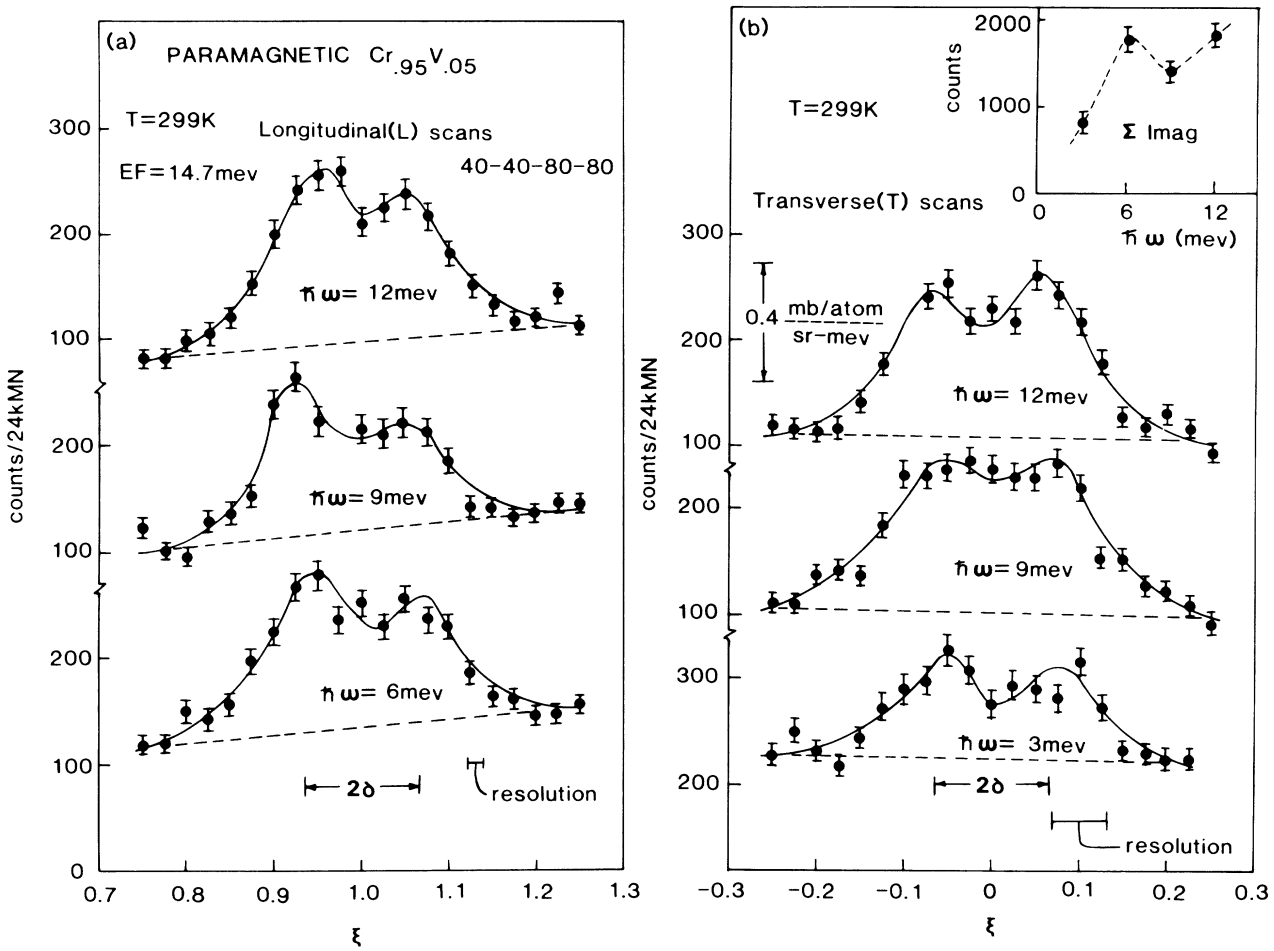

FIG. 2. (a) Longitudinal constant-ΔE scans in $Cr_{0.95}V_{0.05}$ at various energy transfers $\hbar\omega$, at temperature $T=299$ K. The scattering vector is $\mathbf{q}=[\xi,0,0]a^*$. (b) Transverse constant-ΔE scans at various energy transfers $\hbar\omega$, at $T=299$ K. The scattering vector is $\mathbf{q}=[1,\xi,0]a^*$. Inset: Integrated magnetic intensity ΣI_{mag} of these scans as a function of excitation energy $\hbar\omega$. The counting time per point is about 8 min.

point, and its spectral width is much smaller. We speculate that the reduction of the CD scattering to near zero in $Cr_{0.95}V_{0.05}$ is due to pinning of the phase of the dynamic SDW's to the lattice via the Coulomb interactions between the V ions and the charge-density wave concomitant with the SDW,[9] or to the direct pinning of the SDW to the Friedel charge oscillations surrounding the V impurity.[10] The incommensurate inelastic scattering that we are seeing in $Cr_{0.95}V_{0.05}$ is too weak to be observable against the strong CD scattering in pure Cr.

We have also carried out experiments on $Cr_{0.995}V_{0.005}$. To a first approximation, there is no CD scattering. The spectral distribution of the scattering of the satellites in $Cr_{0.995}V_{0.005}$ has a Lorentzian width of $\Gamma \approx 7$ meV. The magnitude of the magnetic scattering cross section is about a factor 10 less than in pure Cr. Thus, in this quite dilute alloy, the static incommensurate SDW state below the Néel point is already foreshadowed in the paramagnetic phase, which is not the case in pure Cr.

The addition of V to Cr evidently changes the nature of the magnetic excitations in the paramagnetic phase, as well as depressing the Néel temperature. This behavior is undoubtedly related to the dramatic changes seen at the Néel transition in the thermophysical properties.[11,12] As little as 0.2 at.% V changes the first-order transition, which is seen in pure Cr as a step in the plot of strain versus temperature, into a quasicontinuous transition.[11] Furthermore, the strong attenuation peak in the longitudinal sound wave propagation seen in pure Cr is greatly reduced in dilute *Cr*V alloys.[12,13] Perhaps the most significant change is the considerably reduced magnitude of the magnetic contribution to the thermal expansivity seen[14] in the paramagnetic state of $Cr_{0.995}V_{0.005}$ compared with pure Cr. From our inelastic neutron-scattering data on the *Cr*V alloys it seems reasonable to conclude that these drastic changes in the thermophysical properties are directly related to the disappearance of the large commensurate-diffuse scattering seen in pure Cr.

The interesting feature of the temperature dependence of the SDW paramagnons in $Cr_{0.95}V_{0.05}$ is that their am-

plitude *increases* with temperature. Similar behavior has been observed in MnSi[15] and YMn_2.[16] It has been suggested that this behavior is related to that predicted for weak itinerant-electron ferromagnets by Moriya and Kawabata,[17] and is a feature of several later calculations.[18] Recent calculations of the many-body enhanced wave-vector-dependent magnetic susceptibility $\chi(\mathbf{q})$ show a singularity at $2k_F$ for Cr but not for Mo, which has a similar Fermi surface.[19] Theoretical results, directly relevant to our measurements of the dynamic susceptibility $\chi(\mathbf{q},\omega)$ at finite temperatures in paramagnetic Cr and *Cr*V alloys, are not yet available.

The paramagnetic excitations, with a very steep (nearly vertical) effective dispersion described here for $Cr_{0.95}V_{0.05}$, have also recently been observed[20] above the Néel temperature in La_2CuO_4, which has been described as a quantum spin fluid. In conclusion, we suggest the likelihood that SDW paramagnons exist in many metals, but have remained undetected for two reasons: (1) Previous work has concentrated on the search for static SDW by elastic neutron scattering, which if it exists at all may be obscured by incoherent nuclear scattering; (2) in most cases, one has no clue where to look. In $Cr_{0.95}V_{0.05}$ the existence of a static SDW in the neighboring pure Cr provides the clue.

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[1]The characteristic feature of the Overhauser static spin-density wave [A. W. Overhauser, Phys. Rev. **128**, 1437 (1962)] is its incommensurability with the lattice, corresponding to a singularity of the wave-vector-dependent susceptibility $\chi(\mathbf{q})$ at a value of $\mathbf{q} \approx 2\mathbf{k}_F$ spanning the Fermi surface. Here, we generalize this concept to the wave-vector- and frequency-dependent susceptibility, $\chi(\mathbf{q},\omega)$.

[2]Paramagnons [the concept, but not the term, first introduced by T. Izuyama, D. J. Kim, and R. Kubo, J. Phys. Soc. Jpn. **18**, 1025 (1963)] are strongly damped spin fluctuations in the paramagnetic state, which renormalize the magnetic susceptibility χ.

[3]For a comprehensive review of the SDW antiferromagnetism in Cr, see E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).

[4]W. C. Koehler, R. M. Moon, A. L. Trego, and A. R. Mackintosh, Phys. Rev. **151**, 405 (1966).

[5]B. H. Grier, G. Shirane, and S. A. Werner, Phys. Rev. B **31**, 2882 (1985).

[6]C. R. Fincher, G. Shirane, and S. A. Werner, Phys. Rev. B **24**, 1312 (1981).

[7]K. R. A. Ziebeck, J. G. Booth, P. J. Brown, H. Capellmann, and J. A. C. Bland, Z. Phys. B **48**, 233 (1982).

[8]A. W. Overhauser, Phys. Rev. B **3**, 3173 (1971), predicted phase fluctuations of charge-density waves, which he termed phasons.

[9]P. B. Littlewood and T. M. Rice, Phys. Rev. Lett. **48**, 44 (1982).

[10]I. Tüttö and A. Zawadowski, Phys. Rev. Lett. **60**, 1442 (1982).

[11]E. Fawcett, R. B. Roberts, A. Day, and G. K. White, Europhys. Lett. **1**, 473 (1986). P. C. de Camargo and E. Fawcett (unpublished) find that, in a better crystal of Cr+0.2 at.% V, the Néel transition is considerably sharper (but still continuous, with no sign of a first-order discontinuity in length) than in Fig. 1(c) of Ref. 10.

[12]de Camargo and Brotzen, Ref. 11.

[13]E. Fawcett, C. S. Ho, and J. M. Perz, Phys. Rev. Lett. A **53** 27 (1975).

[14]E. Fawcett, A. B. Kaiser, and G. K. White, Phys. Rev. B **34**, 6248 (1986). [See also Fig. 1 of E. Fawcett, A. B. Kaiser, and G. K. White, J. Magn. Magn. Mater. **54-57**, 1021 (1986).

[15]Y. Ishikawa, Y. Noda, Y. J. Uemura, C. F. Majkrzak, and G. Shirane, Phys. Rev. B **31**, 5884 (1985).

[16]J. Deportes, B. Ouladdiaf, and K. R. A. Ziebeck, J. Magn. Magn. Mater. **70**, 14 (1987); T. Freltoft, P. Böni, G. Shirane, and K. Motoya, Phys. Rev. B **37**, 3454 (1988).

[17]T. Moriya and A. Kawabata, J. Phys. Soc. Jpn. **34**, 639 (1973).

[18]S. N. Evangelou, H. Hasegawa, and D. M. Edwards, J. Phys. F **12**, 2035 (1982); M. Shimizu, J. Inoue, U. Ohta, and K. Niwa, Physica (Amsterdam) **119B**, 3 (1983); G. G. Lonzarich, J. Magn. Magn. Mater. **54-57**, 612 (1986); H. Hasegawa, J. Phys. F **16**, 1555 (1986).

[19]Y. Z. Zhao, J. L. Fry, P. C. Pattnaik, and K. Schwartzman, to be published.

[20]G. Shirane, Y. Endoh R. J. Birgenau, M. A. Kastner, Y. Hidaka, M. Oda, M. Suzuki, and T. Murabami, Phys. Rev. Lett. **59**, 1613 (1987).