The strongly enhanced magnetic excitations near the quantum critical point of Cr$_{1-x}$V$_x$ and why strong exchange enhancement need not imply heavy fermion behavior

S. M. Hayden$^1$, R. Double$^1$, G. Aeppli$^2$, T. G. Perring$^3$, E. Fawcett$^4$

$^1$H. H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol BS8 1TL, United Kingdom
$^2$NEC Research Institute, 4 Independence Way, Princeton, NJ 08540
$^3$ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, OX11 0QX, United Kingdom
$^4$Department of Physics, University of Toronto, Toronto, M5S 1A7, Canada

(18 November 1999)

Inelastic neutron scattering reveals strong spin fluctuations with energies as high as 0.4eV in the nearly antiferromagnetic metal Cr$_{0.95}$V$_{0.05}$. The magnetic response is well described by a modified Millis-Monien-Pines function. From the low-energy response, we deduce a large exchange enhancement, more than an order of magnitude larger than the corresponding enhancement of the low-temperature electronic heat capacity $\gamma T$. A scaling relationship between $\gamma$ and the inverse of the wavevector-averaged spin relaxation rate $\Gamma_{\text{ave}}$ is demonstrated for a number of magnetically correlated metals.

Many metals close to a magnetic instability at low temperatures display novel behavior in their physical properties, for example a linear temperature dependence of the resistivity. Notable examples include the lamellar CuO systems, non-Fermi liquid systems, heavy fermions and nearly ferromagnetic metals. The proximity of an antiferromagnetic phase is also widely believed to be related to the occurrence of some forms of superconductivity. Clearly, the magnetic excitations are largely responsible for the properties of such materials. In this paper, we study the magnetic excitations near the quantum critical point (QCP) of the structurally simple (body centered cubic) 3d metal Cr$_{1-x}$V$_x$. For $x \leq 0.35$, Cr$_{1-x}$V$_x$ displays antiferromagnetic order in the form of an incommensurate spin density wave as in pure chromium. The point ($x=0.35$,$T=0$) in the $x-T$ plane (see Fig. 1(a)) is a QCP where a phase transition (the ordering of 3d moments) occurs at zero temperature.

Specifically, we studied the nearly antiferromagnetic composition Cr$_{0.95}$V$_{0.05}$. Strong magnetic excitations were observed: in common with 4f and 5f materials near quantum criticality. However, in contrast, the excitations in Cr$_{0.95}$V$_{0.05}$ are strong only in a small portion of reciprocal space. Other findings of the present study are: (i) magnetic excitations exist with energies comparable to those of the highest frequency spin waves in strong ferromagnets such as iron and nickel; (ii) a simple phenomenological response function with four parameters describes the magnetic response over an extremely wide (0.004-0.4eV) energy range; (iii) Cr$_{0.95}$V$_{0.05}$ shows a large exchange enhancement of 28 ± 4; (iv) there is a universal relationship between the Brillouin zone (BZ) averaged spin relaxation rate and the electronic heat capacity for Cr$_{0.95}$V$_{0.05}$ and other magnetically correlated metals; (v) the low effective mass of the quasiparticles in Cr$_{0.95}$V$_{0.05}$ which coexists with the high exchange enhancement is due to the small phase space occupied by the exchange enhanced fluctuations.

Reactor-based neutron scattering measurements have demonstrated incommensurate magnetic correlations in Cr$_{0.95}$V$_{0.05}$ for thermal energies. In particular, the low frequency magnetic response peaks at six symmetry-equivalent positions near the H or (100)-type point of the BZ. By using the HET and MARI spectrometers at the ISIS spallation source, we have extended the energy range by almost an order of magnitude. Absolute intensities were obtained via normalization to measured incoherent scattering from a vanadium standard and coherent phonon scattering from the sample. The accuracy is about ±10%. Our sample, from the Materials Preparation Center of Ames Laboratory, was a 47.9 g arc-melted single crystal. The lattice constant at $T=2$ K was $a=2.895$ Å. Elastic scans performed using a cold-source three-axis spectrometer ($k_i=1.5$ Å$^{-1}$) revealed no antiferromagnetic order for temperatures as low as $T=2$ K.

Neutron scattering measures the imaginary part of the generalized magnetic susceptibility $\chi''(Q,\omega)$, the cross-section for a paramagnet such as Cr$_{0.95}$V$_{0.05}$ is,

$$\frac{d^2\sigma}{d\Omega dE} = (\gamma r_e)^2 \frac{k_f}{k_i} |F(Q)|^2 \left(\frac{2/\pi g^2\mu_B^2}{1 - \exp(-\hbar\omega/kT)}\right) \times \chi''(Q,\hbar\omega),$$

(1)

where $(\gamma r_e)^2=0.2905$ barn sr$^{-1}$, $k_i$ and $k_f$ are the incident and final neutron wavevectors and $|F(Q)|^2$ is the magnetic form factor. We label wavevectors by their positions in reciprocal space $Q = (h,k,l)$.

We first characterized the inelastic scattering at lower $\omega$. Previous measurements have established that for $\hbar\omega \approx 10$ meV, the magnetic response is peaked at the six equivalent positions $Q_d = (1 \pm \delta,0,0), (1,\pm\delta,0)$ and $\ldots$
under these conditions are shown in Fig. 1(c). The re-
for $\bar{\mathbf{h}}\omega \approx 25 \text{ meV}$ as shown in Fig. 1(b). Data collected
investigated. For all energies the response is
high energy transfer increases the scattering broadens in
the excitations probably exist up to even higher
wavevector and only one broad peak is observed at the
energy, we are able to cut through the ‘column’ of scattering
depicted in Fig. 1(b) at different energy transfers. The left
and right panels show cuts in two perpendicular reciprocal
space directions. Open circles are a background measured
at the same $\omega$ and $|\mathbf{Q}|$ but away from the H position
and the shaded area is the magnetic signal. The incident energies
were (from bottom) $E_i=35, 110, 705, 860 \text{ meV}$. Solid lines are
resolution-corrected fits of Eq. (4) to the data.

A number of response functions have been used to de-
scribe nearly antiferromagnetic metals [12–17]. Following
Zha et al. [16] for La$_{2-x}$Sr$_x$CuO$_4$, we use the function
\[
\chi(Q, \omega) = \sum_{Q_\delta} \chi_\delta \left[ 1 + \frac{\mathbf{Q} - \mathbf{Q}_\delta^2}{\kappa_0^2} - i \frac{\omega}{\omega_{\text{SF}}} \right]^{-1},
\]
where the sum is over the six $Q_\delta$ positions surrounding
the H point of the BZ. Eq. (2) may be justified by expanding the Lindhard functions near the Fermi energy [12] and
thus should describe a Fermi liquid. It also has a more general phenological justification in that it represents the
first symmetry-allowed terms in a power series expansion of $\chi^{-1}(Q, \omega)$. The observed scattering is related to the
imaginary part of $\chi$, which is,
\[
\chi''(Q, \omega) = \sum_{Q_\delta} \frac{\chi_\delta r_0^4 [\omega/\omega_{\text{SF}}]}{[\kappa_0^2 + (\mathbf{Q} - \mathbf{Q}_\delta)^2]^2 + [\omega/\omega_{\text{SF}}]^2 \kappa_0^4},
\]
The physical meaning of the parameters in Eq. (3) is now
clear. $\chi_\delta$ determines the overall amplitude of the re-
sponse. In the low-frequency limit $\omega \ll \omega_{\text{SF}}$, the position

FIG. 1. (a) The magnetic phase diagram of the Cr$_{1-x}$V$_x$
system showing the incommensurate antiferromagnet (AF)
and paramagnetic (P) phases. (b) Schematic representation
of the dynamic susceptibility $\chi''(Q, \omega)$ (Eq. 2) of Cr$_{0.95}$V$_{0.05}$
with experimentally determined parameters. For each energy,
a contour at $1/2$ the maximum value of $\chi''(Q, \omega)$ is drawn,
the shading represents the maximum value. For $E_i=35$
meV, $\chi''(Q, \omega)$ is sampled on the grey surface. (c) Magnetic
fluctuations near (100) in Cr$_{0.95}$V$_{0.05}$ ($T=12 \text{ K}$) measured
using the HET spectrometer.

FIG. 2. Magnetic scattering near the (100) and (210)
reciprocal space directions. Open circles are a background measured
at the same $\omega$ and $|\mathbf{Q}|$ but away from the H position
and the shaded area is the magnetic signal. The incident energies
were (from bottom) $E_i=35, 110, 705, 860 \text{ meV}$. Solid lines are
resolution-corrected fits of Eq. (4) to the data.
and sharpness of the peaks in wavevector are determined by $\delta$ and $\kappa_0$ respectively. We find these parameters to be $\delta=0.08\pm0.01$ r.l.u. and $\kappa_0=0.11\pm0.01$ A$^{-1}$ from fitting low-frequency ($h\omega=4$ meV) data collected on a cold-source triple axis spectrometer. Finally, the frequency dependence of the response i.e. how the peak widths and amplitudes change with frequency is controlled by the term $(\omega/\omega_{SF})^2\kappa_0$, where $\omega_{SF}$ is the characteristic frequency over which changes occur.

To test whether Eq. 3 describes the evolution of our data with $\omega$, we fitted cuts at each $\omega$ using the general form

$$\chi''(\mathbf{Q},\omega) = \sum_{\mathbf{Q}_3} \frac{\chi'_0(\omega)[\kappa_3^2 + \kappa_4^2(\omega)]}{[\kappa_0^2 + (\mathbf{Q} - \mathbf{Q}_3)^2]^2 + \kappa_4^2(\omega)},$$  \tag{4}

where the fitted $\omega$-dependent parameters $\chi'_0(\omega)$ and $\kappa_4(\omega)$ control the height and sharpness of the incommensurate peaks respectively. The parameters $\delta$ and $\kappa_0$ were fixed at the values given above. If Eq. 3 describes the response, comparing Eq. 4 and Eq. 5 predicts that the fitted parameters will vary as,

$$\kappa_3^2(\omega) = \frac{\chi_0[\omega/\omega_{SF}]}{1 + [\omega^2/\omega_{SF}^2]},$$  \tag{5}

and

$$\kappa_4^2(\omega) = \frac{\omega}{\omega_{SF}} \kappa_3^2.$$  \tag{6}

The solid lines in Fig. 3 correspond to resolution-corrected fits of Eqs. 4-6 for different $\omega$. Fig. 3(a) and (b) show the resulting $\chi'_0(\omega)$ and $\kappa_4^2(\omega)$ values for all data collected. The solid line in Fig. 3(a) is a fit of Eq. 4 which yields values for the spin fluctuation energy and amplitude parameters of $h\omega_{SF}=88\pm10$ meV and $\chi_0=45\pm3 \mu_0^2$ eV$^{-1}$ f.u.$^{-1}$ respectively. Fig. 3(b) shows that the width parameter $\kappa_3^2$ displays an approximately linear variation with $\omega$ as suggested by Eq. 5. The gradient of the fitted line $\kappa_3^2/\omega_{SF}=0.118\pm0.006$ A$^{-2}$ eV$^{-1}$ gives a second estimate of $h\omega_{SF}=102\pm24$ meV. Obtaining two indistinguishable estimates of $\omega_{SF}$ in this way demonstrates that Eq. 4 provides a good description of our data: the $\omega$ dependence of the the amplitude and sharpness of the peaks are controlled by the parameter $\omega_{SF}$.

The starting point for models of the dynamic response of chromium and its alloys is the Fermi surface. It is believed that the peak in the dynamic susceptibility is due to the imperfect nesting of an electron ‘jack’ and a larger hole ‘octahedron’. Staunton et al. have recently performed $ab$-initio calculations of the spin susceptibility for Cr$_{0.95}$V$_{0.05}$. To compare our results with these and other theoretical models, we have evaluated the $\omega$ dependence of $\chi''(\mathbf{Q},\omega)$ for each $\omega$ probed assuming Eq. 4. The resulting response is shown in Fig. 3(c).

$\chi''(\mathbf{Q},\omega)$ differs from $\chi'_0(\omega)$ because Eq. 3 sums contributions centered at six equivalent and nearby wavevectors $\mathbf{Q}_3$. The dashed line in Fig. 3(c) shows the non-interacting susceptibility $\chi'_0(\mathbf{Q},\omega)$ calculated from the band structure and neglecting the exchange interaction i.e. the bare Lindhard function [15]. Within a simple RPA model, the interacting susceptibility $\chi(\mathbf{Q},\omega)$ is given by $\chi^{-1}(\mathbf{Q},\omega) = \chi_0^{-1}(\mathbf{Q},\omega) - \lambda$, where $\lambda$ is the mean field parameter describing the Coulomb interaction. In the low $\omega$ limit $\chi''(\mathbf{Q},\omega) \propto \omega$ and,

$$\chi''(\mathbf{Q},\omega) = \frac{\chi_0''(\mathbf{Q},\omega)}{[1 - \lambda \chi_0''(\mathbf{Q},\omega)]^2}. \tag{7}$$

Comparing the initial gradients of the two lines in Fig. 3(c), we estimate the exchange enhancement factor $[1 - \lambda \chi_0''(\mathbf{Q},\omega)]^{-1} = 28\pm4$, demonstrating that Cr$_{0.95}$V$_{0.05}$ is indeed very strongly exchange enhanced.

Many systems displaying strong magnetic fluctuations show dramatically renormalized thermodynamic properties. For example, heavy fermions exhibit a very large electronic linear heat capacity at low temperature. In spite of its large exchange enhancement, Cr$_{0.95}$V$_{0.05}$ has a relatively small electronic specific heat $\gamma T$, $\gamma=2$ mJK$^{-2}$ [15]. To understand why the quasiparticles in Cr$_{0.95}$V$_{0.05}$ are not heavier, we must estimate the magnetic contribution to $\gamma$. If the excitations can be described as a set of overdamped oscillators with wavevector-dependent relax-
estimates of the uncertainty in $\Gamma$ have been computed from data in Refs. [23], horizontal bars are estimates of the uncertainty in $\Gamma_{\text{ave}}$. The dotted line is Eq. 8. The coefficient $\gamma$ is plotted against $\omega$. 

FIG. 4. The coefficient $\gamma$ of the low temperature electronic specific heat $\gamma T$ is plotted against the wavevector-averaged spin relaxation rate $\Gamma_{\text{ave}}$, where $\Gamma_{\text{ave}}^{-1} = \langle \Gamma^{-1}(Q) \rangle_{\text{BZ}}$. $\Gamma_{\text{ave}}$ has been computed from data in Refs. [23]. horizontal bars are estimates of the uncertainty in $\Gamma_{\text{ave}}$. The dotted line is Eq. 8.

The universal relationship Eq. 8 holds across widely differing systems such as 3$d$ transition metals, their oxides, and heavy fermion compounds, demonstrating that the spin channel dominates the electronic entropy in all of these systems.

One might view the materials in Fig. 4 as being close to a QCP. Each of the materials has 'soft' magnetic excitations in some region of reciprocal space. In spite of the large enhancement in Cr$_{0.95}$V$_{0.05}$, the magnetic fluctuations in a relatively small portion of reciprocal space (where $\Gamma$ is small) contribute to $\gamma$. In contrast, the heavy fermion systems have large $\gamma$ because the spin fluctuations are soft (have low $\omega$) over larger portions of reciprocal space. For predominately antiferromagnetic fluctuations, $\gamma$ scales roughly as $\kappa_0^2/\omega_{\text{SF}}$, where $d$ is the system dimensionality. Thus, to raise $\gamma$, it is advantageous to lower $d$ and $\omega_{\text{SF}}$ and increase $\kappa_0$. Heavy fermion systems have larger $\kappa_0$ and smaller $\omega_{\text{SF}}$ than Cr$_{0.95}$V$_{0.05}$ and hence larger $\gamma$ values.

In summary, we have measured the high-frequency dynamic magnetic response of the paramagnetic alloy Cr$_{0.95}$V$_{0.05}$ which is close to incommensurate magnetic order and has a large exchange enhancement. We observe strong magnetic correlations at epithermal energies up to 400 meV. The observed response can be described by a remarkably-simple modified-MMP function where the $\omega$ dependence of the response is controlled by a single parameter. We have computed the BZ average of the magnetic relaxation rate for Cr$_{0.95}$V$_{0.05}$ and other materials close to magnetic order (or a QCP), this demonstrates the relationship between this quantity and the low-temperature quasiparticle specific heat, and also account for why Cr$_{0.95}$V$_{0.05}$ in spite of its large exchange enhancement, is not a heavy fermion system.

We grateful for helpful discussions with P. W. Michell, J. Lowden, R. Fishman, J. Staunton and O. Stockert.

[1] M. A. Kastner et al., Rev. Mod. Phys. 70 897 (1998).
[2] Non-Fermi Liquid Behaviour in Metals (Santa Barbara, 17-21 June 1996), J. Phys. C 8, 9675-10148 (1996).
[3] G. H. Lander and G. Aeppli, J Mag. Mag. Mat. 100, 151 (1991).
[4] N. R. Bernhoeft et al., Phys. Rev. Lett. 62, 657 (1989).
[5] N. D. Mathur et al., Nature 394, 39 (1998).
[6] E. Fawcett, Rev. Mod. Phys. 66, 209 (1988) and E. Fawcett et al., ibid 66, 25 (1994).
[7] D. McK. Paul et al., Phys. Rev. B 38, 580 (1988).
[8] H. A. Mook and D. M. Paul, Phys. Rev. Lett. 54, 227 (1985).
[9] E. Fawcett et al., Phys. Rev. Lett. 61, 558 (1988); S. A. Werner et al., J. Appl. Phys. 73, 6454 (1993).
[10] S. M. Hayden et al., Physica B 241-243, 241 (1998); ibid 237, 421 (1997).
[11] R. Double PhD Thesis, University of Bristol (1998).
[12] T. Moriya, Phys. Rev. Lett 24, 1433 (1970).
[13] H. Sato and K. Maki, Int. J. Magnetism 6, 183 (1974).
[14] A. J. Millis, H. Monien, and D. Pines, Phys. Rev. B 42, 167 (1990).
[15] D. R. Noakes et al., Phys. Rev. Lett. 65, 369 (1990).
[16] Y. Zha, V. Barzykin, and D. Pines, Phys. Rev. B 54, 7561 (1996).
[17] G. Aeppli et al., Science 278, 1432 (1997).
[18] J. B. Staunton et al., Phys. Rev. Lett. 83 3340 (1999); private communication.
[19] F. Heiniger Phys. kondens. Materie 5, 285 (1966).
[20] D. M. Edwards and G. G. Lonzarich, Phil. Mag. B 65, 1185 (1992).
[21] T. Moriya and T. Takimoto, J. Phys. Soc. Japn. 64, 960 (1995).
[22] $\Gamma$ is defined from the response of an overdamped oscillator $\chi''(Q, \omega) \propto \omega/\left(\omega^2 + \Gamma^2\right)$.
[23] L. P. Regnault et al., J. Mag. Mag. Mat. 63-64, 289 (1987) (CeCu$_6$); Ref. [4] and N. R. Bernhoeft and G. G. Lonzarich, J. Phys.: Cond. Mat. 7, 7325 (1995) (UPt$_3$); N. R. Bernhoeft et al., Phys. Rev. Lett. 81, 4244 (1998) (UPd$_2$Al$_3$); Ref. [4] (Ni$_3$Ga); Ref. [4] (Pd); Ref. [4] (La$_{1-x}$Sr$_x$CuO$_4$).