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1993 J. Phys.: Condens. Matter 5 3959

(http://iopscience.iop.org/0953-8984/5/23/022)

J. Phys.: Condens. Matter 5 (1993) 3959–3964. Printed in the UK

Dynamics of paramagnetic chromium alloys

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Received 29 January 1993

Abstract. Despite the absence of a static spin-density wave (SDW), the cross section of paramagnetic chromium alloys still peaks near the wavevectors of the SDW in pure chromium. An analysis of the paramagnetic fluctuations in doped chromium reveals that the cross section vanishes at zero temperature and zero frequency, in agreement with recent experiments by Fawcett *et al*. We also predict that the peak elastic cross section will reach a maximum at a temperature which increases with the impurity concentration and that impurity scattering will eventually broaden the sidepeaks into a single central peak.

Doping with even a small fraction of impurities has a dramatic effect on the magnetic properties of chromium. Pure chromium experiences a weakly first-order phase transition [1–4] into a spin-density wave (SDW) state [5] at the Néel temperature $T_N \simeq 310$ K (26.7 meV). While adding 0.2% vanadium impurities drives the phase transition to second order [3], adding 4% vanadium destroys [6,7] the SDW and drives the Néel temperature to zero. But despite the absence of an SDW at any temperature in paramagnetic CrV alloys, the neutron-scattering cross section σ still peaks near the wavevector of the SDW in pure chromium. While some theorists [8] have speculated that magnons may continue to exist above the Néel temperature in itinerant antiferromagnets, we argue that the peaks in the paramagnetic cross section of chromium alloys are due to thermal fluctuations. By evaluating the spin susceptibility in the paramagnetic regime with $T_N = 0$, we find that that the elastic cross section vanishes at zero temperature, in agreement with the measurements of Fawcett *et al* [7]. We also predict the behaviour of the cross section as a function of frequency and temperature.

The band structure of pure chromium [9] consists of an electron 'jack' (surface a) and a slightly larger hole 'octahedron' (surface b) which are imperfectly nested by the wavevector $\boldsymbol{Q} = 2\pi(1-\delta)\hat{\boldsymbol{z}}/a$, where a is the lattice constant of the BCC lattice and $\delta \simeq 0.04$. Due to their symmetry, the Fermi surfaces are also imperfectly nested by the wavevector $\boldsymbol{G} - \boldsymbol{Q}$, where $\boldsymbol{G} = (4\pi/a)\hat{\boldsymbol{z}}$ is a reciprocal lattice vector. Because vanadium has fewer conduction electrons than chromium, doping with vanadium increases the size of the hole surface and the magnitude of the mismatch δ. Doping with manganese, on the other hand, increases the size of the electron surface and suppresses δ.

The weak first-order transition in pure chromium was clearly observed by Arrott and co-workers [1], who measured a discontinous change at the Néel temperature in the neutron scattering intensities at a wavevector very close to $\boldsymbol{Q}$. Because $\boldsymbol{Q}$ differs from $\boldsymbol{G}/2$, the SDW is incommensurate with the lattice and the two sides of the hole Fermi surface are not equivalent. So the band structure of chromium actually consists of one electron and two

0953-8984/93/233959+06$07.50

hole surfaces. In pure chromium, the energy mismatch between the b± Fermi surfaces, when translated by $\pm\boldsymbol{Q}$, is given by [9] $z_0 \simeq 500$ meV.

While the Coulomb attraction λ between the electron and hole surfaces is responsible for the SDW, the Coulomb repulsion λ' between the holes on the b+ and b− Fermi surfaces is responsible for the appearance of a charge-density wave (CDW) [10] with twice the wavevector of the SDW. As discussed in a previous paper [11], the first-order phase transition is induced by the proximity to a CDW instability when $\lambda' = 0.5$. Since impurities scatter electrons and holes with opposite signs [12], impurity scattering competes with the electron–hole Coulomb interaction λ and suppresses both the SDW order parameter g and the CDW order parameter $\partial \propto g^2$. We previously demonstrated [11] that the first-order phase transition is destroyed by approximately 0.15% vanadium concentration, in agreement with neutron scattering [3] and strain [4] measurements.

When chromium is doped with more than 4% vanadium, the SDW is destroyed and the Néel temperature vanishes [7]. But despite the absence of an SDW at all temperatures, the neutron scattering cross section still contains peaks near the nesting wavevector $\boldsymbol{Q}$. The paramagnetic cross sections are, however, about 100 times smaller than the cross section just above the Néel temperature of pure chromium [13]. Fawcett *et al* [7] also find that the elastic scattering cross section is negligible at $T = 9$ K.

To calculate the neutron scattering cross section, we construct the Matsubara spin susceptibility

$$\chi_{\alpha\beta}(\boldsymbol{q}, \omega_n) = \frac{1}{4N}\sum_{k,k'}\int_0^\beta d\tau\, e^{i\omega_n\tau}\langle T_\tau \Psi^\dagger(\boldsymbol{k}+\boldsymbol{q}, \tau)\cdot\boldsymbol{\Lambda}_\beta\cdot\Psi(\boldsymbol{k}, \tau)\Psi^\dagger(\boldsymbol{k}', 0)\cdot\boldsymbol{\Lambda}_\alpha\cdot\Psi(\boldsymbol{k}'+\boldsymbol{q}, 0)\rangle \tag{1}$$

where

$$\boldsymbol{\Lambda}_\beta = \begin{pmatrix} \sigma_\beta & \sigma_\beta & \sigma_\beta \\ \sigma_\beta & \sigma_\beta & \sigma_\beta \\ \sigma_\beta & \sigma_\beta & \sigma_\beta \end{pmatrix} \tag{2}$$

σ_β are the Pauli matrices in spin space, $\omega_n = 2n\pi T$ are the boson Matsubara frequencies, and $\Psi(\boldsymbol{k}, \tau)$ is the six-dimensional Fermi operator in spin and band space.

We calculate the susceptibility for paramagnetic chromium alloys by incorporating the Coulomb and scattering corrections to the Hartree–Fock susceptibility in the usual way [14]. The neutron scattering cross section is then proportional to the imaginary transverse susceptibility

$$\mathrm{Im}(\chi_{+-}(\boldsymbol{q}, \omega)) = 2\Phi_2(\boldsymbol{q}, \omega)/[(1 - \nu\Phi_1(\boldsymbol{q}, \omega))^2 + (\nu\Phi_2(\boldsymbol{q}, \omega))^2] \tag{3}$$

where $\nu = N(0)\lambda$ is the electron–hole Coulomb constant and $\chi_{+-}(\boldsymbol{q}, \omega)$ is given by the analytic continuation of equation (1). The functions $\Phi_1(\boldsymbol{q}, \omega)$ and $\Phi_2(\boldsymbol{q}, \omega)$ are the real and imaginary parts of [14]

$$\Phi(\boldsymbol{q}, \omega) = \int\frac{dz_1}{2\pi}\int\frac{dz_2}{2\pi}a(z_1, z_2)\frac{f(z_2) - f(z_1)}{z_1 - z_2 + \omega + i\varepsilon} \tag{4}$$

where $a(z_1, z_2)$ is the spectral density [15]

$$a(z_1, z_2) = [4\pi\Gamma N(0)/D(z_1 + z_2)]\{(z_1 + z_2 - \eta)(z_1 + z_2 + \eta - z_0) - 2\Gamma^2 - 3(z_1 + z_2 - z_0/2)^2\} \tag{5}$$

$$D(z) = [(z - \eta)(z + \eta - z_0) - 2\Gamma^2]^2 + 9(z - z_0/2)^2\Gamma^2 \tag{6}$$

$f(z) = 1/(1+\exp(\beta z))$ is the Fermi function, and $N(0)$ is the density of states per spin. The damping energy $\Gamma = \hbar/\tau$ is inversely related to the scattering lifetime τ of the electrons. With q parallel to the nesting wavevector $Q = Q\hat{z}$, $\eta = v_F(q-Q)/\sqrt{3}$ gives the deviation of the wavevector q from Q. Because the hole–hole Coulomb interaction λ' does not contribute above T_N, our results are formally identical to those of Sato and Maki [16], who employed a two-band model.

Since $\Phi_2(q, 0) = 0$, the SDW wavevectors and Néel temperature are given by the zeros of $D(T, q) = 1 - \nu\Phi_1(q, 0)$. When Γ exceeds a critical value denoted by Γ_{cr}, $T_N = 0$ and $D(T, q)$ does not vanish at any temperature or wavevector. In the limit of zero frequency but finite temperature ($\beta\omega \to 0$), the elastic cross section is proportional to

$$\lim_{\omega\to 0} \frac{\Phi_2(q,\omega)}{1-e^{-\beta\omega}} = \frac{T}{2}\int_{-\infty}^{\infty} \frac{dz}{2\pi} a(z,z) f'(z). \tag{7}$$

Now in the limit of zero temperature, $f'(z) \to -\delta(z)$ and the elastic cross section must vanish, in agreement with the observation of Fawcett *et al* [7].

Surprisingly, we find that for finite temperatures, the elastic cross section does not peak at the wavevectors Q and $G - Q$. In fact, when $T_N > 0$, the wavector Q' of the SDW is actually given by $Q' = Q + K$, where

$$K = \frac{2\pi}{a}\frac{2\zeta}{z_0}\delta\hat{z}. \tag{8}$$

The shifted hole and electron Fermi surfaces are sketched in figure 1. In order to maximize the nesting on both sides of the Fermi surface, the energy mismatch between the b± Fermi surfaces, now translated by $\pm Q'$ instead of $\pm Q$, is reduced from z_0 to $z_0 - \zeta$. The resulting band structure is sketched in figure 2. For large z_0, ζ is of the order of T_N and K is of the order of GT_N/z_0.

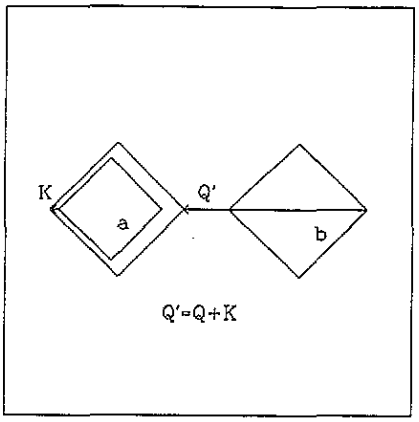

Figure 1. Schematic drawing of the hole and electron bands in chromium, which are imperfectly nested by the wavevector Q'.

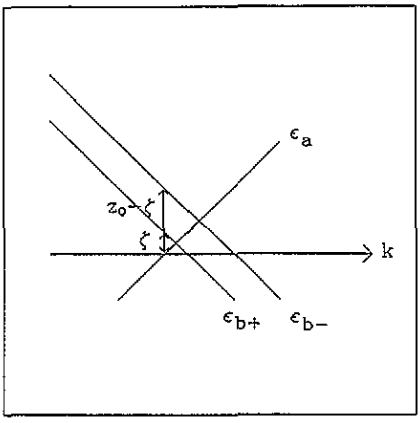

Figure 2. The band structure of chromium near the Fermi energy when the b± surfaces are translated by $\pm Q'$.

This can be easily understood by considering the nesting of the Fermi surfaces for $k_z > 0$. Due to the displacement K of the Fermi surfaces, the condensation energy of

the b− and a Fermi surfaces increases by an amount of order T_N. Since the condensation energy of the b+ and a surfaces is maximized when $\zeta = 0$, the shift in the Fermi surfaces by $K \simeq T_N/az_0$ lowers the b + a condensation energy by an amount of the order of T_N^2/z_0. Consequently, the shift in the SDW wavevectors generates a net increase in the condensation energy and Néel temperature.

As z_0 increases, the Néel temperature $T_N(z_0)$ decreases monotonically and approaches a non-zero limit as $z_0 \to \infty$. The ratio $2\zeta/z_0$ is also a decreasing function of z_0 which vanishes as $z_0 \to \infty$ and equals 1 when $z_0 < z_0^* \simeq 2\pi T_N(z_0^*)$. When $\Gamma = 0$, z_0^* is approximately 360 meV. Below the triple point z_0^*, the SDW is commensurate with $Q' = G/2$. As z_0 decreases below z_0^*, the electron Fermi surface continues to grow and the Néel temperature continues to increase. So in agreement with measurements by Koehler *et al* [6], CrMn alloys become commensurate with a Néel temperature which is far smaller than its maximal value achieved when $z_0 = 0$.

As the electron-scattering energy Γ increases, both z_0^* and ζ decrease. Hence, damping favours the optimization of nesting on one side of the hole Fermi surface at the expense of the other, rather than the compromise achieved when $\zeta = z_0/2$. So electron scattering causes the side peaks in the neutron scattering cross section at Q' and $G - Q'$ to move *further apart*. As Γ increases, z_0^* decreases monotonically to a minimum value of about 185 meV. When z_0 is smaller than this minimum value, the SDW is commensurate for all values of Γ below Γ_{cr}.

Despite the absence of a static SDW in paramagnetic CrV alloys, the cross section still peaks near the wavevectors which minimize $D(T, q)$. If $2\pi T/z_0 \ll 1$, both the elastic and inelastic cross sections will peak very close to Q and $G - Q$. But as the temperature increases, the side peaks shift closer together towards $G/2$. If $2\pi T/z_0$ is larger than about 1, then the side peaks will collapse into a single central peak. In the inset to figures 3 and 4, we plot the paramagnetic cross section versus $q' = qa/2\pi$ for $T = 5$ meV, $z_0 = 800$ meV, and $\omega = 0$ or 12.39 meV (3 THz). With these parameters, $2\pi T/z_0 \ll 1$ and the cross sections peak very close to Q and $G - Q$. So as conjectured by Fawcett *et al* [7], the SDW paramagnons in paramagnetic CrV alloys are very similar to the incommensurate paramagnons in pure chromium.

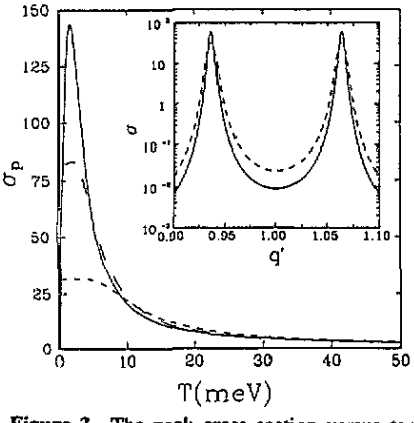

Figure 3. The peak cross section versus temperature for $q = Q$, $\Gamma/\Gamma_{cr} = 1.025$, and $\omega = 0$ (full curve), 4.13 meV (long dashes), or 12.39 meV (short dashes). Inset are the cross sections for $T = 5$ meV (58 K), and $\omega = 0$ (full curve) or 12.39 meV (broken curve).

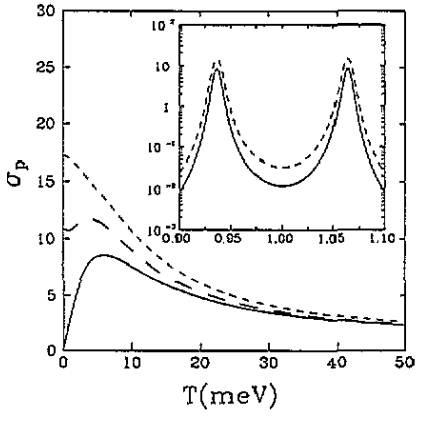

Figure 4. Same as figure 3 but $\Gamma/\Gamma_{cr} = 1.456$.

However, the effect of electron scattering is very different above and below Γ_{cr}. Below Γ_{cr}, damping suppresses the SDW wavevector Q' and causes the side peaks to move further apart; above Γ_{cr}, damping broadens the side peaks until only a central peak at $G/2$ remains. When z_0 lies between 180 and 360 meV, damping below Γ_{cr} will split the central peak into side peaks. Above Γ_{cr}, damping will eventually broaden the side peaks into a central peak. If $z_0 > 360$ meV, electron scattering will first drive the side peaks apart and then meld them together. It would be interesting to test these predictions on CrMo or CrW alloys, so that z_0 does not change with the impurity concentration.

In the limit of large temperatures, $f'(z) \simeq -\beta \exp(-\beta z)$ and the elastic cross section must decrease. So the peaks in the elastic cross section will reach a maximum value at an intermediate temperature $\bar{T}$ that depends on T_N and Γ. When $\Gamma = \Gamma_{cr}$, the denominator $D(T, q)$ will vanish at $T = 0$ and $q \simeq Q$. Consequently, the peaks in the elastic cross section will diverge at zero temperature and $\bar{T}$ must vanish as Γ approaches Γ_{cr}. For small $\Gamma - \Gamma_{cr}$, we believe that $\bar{T} \simeq \Gamma - \Gamma_{cr}$. Since Γ is a linear function of impurity concentration n_i, we expect that $\bar{T}$ also increases linearly with n_i above Γ_{cr}.

In figures 3 and 4, we plot the peak cross section σ_p versus temperature for $q = Q$ and $\Gamma/\Gamma_{cr} = 1.058$ (figure 3) or $\Gamma/\Gamma_{cr} = 1.411$ (figure 4). In agreement with Fawcett *et al* [7], the paramagnetic cross section is roughly 100 times smaller than the cross section above T_N in pure chromium. The peaks in the elastic cross section reach a maximum value which increases in size as Γ approaches Γ_{cr}. In figure 3, σ_p reaches a very large value at $\bar{T} \simeq 1.6$ meV (18.5 K). For the larger value of damping in figure 4, σ_p attains a smaller maximum value at $\bar{T} \simeq 6$ meV (70 K). While σ_p is a monotonically increasing function of frequency for the larger damping in figure 4, σ_p is not a monotonic function of frequency in figure 3.

As the frequency increases from 0, σ_p changes from a non-monotonic function of temperature with a single maximum at $\bar{T}$ to a monotonically decreasing function of temperature. At the intermediate frequency of 1 THz, the maximum of σ_p has moved to a lower temperature and σ_p initially decreases with temperature. So σ_p now contains a minimum at a very small temperature of about 1 meV. This minimum gradually disappears at higher frequencies.

Unfortunately, the measurements of Fawcett *et al* [7] are confined to the temperatures 9 and 299 K. Since the Néel temperature is destroyed by 4% vanadium doping, we may assume that $\Gamma \simeq 1.25\Gamma_{cr}$ in the $Cr_{0.95}V_{0.05}$ sample measured by Fawcett *et al*. In agreement with our results, the measured cross sections increase with frequency. Due to the limited temperature range of their measurements, however, we cannot verify the other predictions of this work.

Clearly, the magnetic properties of chromium are rather complex, even in the paramagnetic regime. In this paper, we demonstrate that thermal fluctuations are responsible for the peaks in the neutron scattering cross section. The elastic cross section vanishes at zero temperature but reaches a maximum at an intermediate temperature which scales with $\Gamma - \Gamma_{cr}$. We hope that this work will encourage a systematic experimental investigation of the properties of paramagnetic chromium alloys.

Acknowledgments

We would like to acknowledge support from the US Department of Energy under contract DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc. and from the EPSCOR (experimental program to stimulate cooperative research) of the National Science Foundation, administered in North Dakota.

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