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Structural order parameter and itinerant electron magnetism in $Cd_2Re_2O_7$

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Abstract

We describe the low-temperature behaviour of the magnetic susceptibility of $Cd_2Re_2O_7$ in terms of a Landau theory of structural phase transitions. We calculate the zero temperature Pauli susceptibility using a tight-binding approach to reveal the mechanism of coupling between the structural order parameter and itinerant magnetism.

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$Cd_2Re_2O_7$ is the only pyrochlore superconductor [1–3]. Upon cooling, $Cd_2Re_2O_7$ undergoes two structural phase transitions (SPTs) well above the very low-temperature superconducting phase transition [4,5]. The first occurs at 200 K and is of second-order, while the other is first-order at 120 K. Neither SPT involves multiplication of the BCC primitive cell. We will refer to the phases as I, II and III, with corresponding space groups $Fd\bar{3}m \rightarrow I\bar{4}m2 \rightarrow I4_122$, respectively. The SPTs are accompanied by anomalous behaviour of physical properties, including the electrical resistivity, magnetic susceptibility, Hall coefficient and thermoelectric power. In this paper, we concentrate on the magnetic properties of $Cd_2Re_2O_7$.

$Cd_2Re_2O_7$ is a paramagnetic metal in the normal state. Above 400 K, the magnetic susceptibility displays approximate Curie–Weiss behaviour with a large, negative Weiss temperature [6]. On cooling, it reaches a broad maximum, and then falls off below 200 K [1,6,7]. No anomaly has been found at the second SPT at 120 K. The kind of anomaly observed at 200 K is often associated with an antiferromagnetic phase transition, but Re nuclear quadrupole resonance experiments find no evidence of magnetic ordering in the low temperature phases [8], thus revealing the itinerant electronic nature of its paramagnetism.

In this article, we show theoretically that the magnetic anomaly at 200 K is due to the electrostatic interaction between itinerant electrons and tetragonal distortions of the pyrochlore lattice. Recently, we found a single two-dimensional order parameter (OP) that describes both SPTs in $Cd_2Re_2O_7$ which corresponds to an instability of the lattice with respect to a long wavelength phonon mode of E_u symmetry [9].

We introduce the two components of the structural OP (η_1, η_2), which are linear combinations of the displacements of four Re atoms, corresponding to one BCC site, from their ideal positions in phase I, (x_m, y_m, z_m), $m = 1, 2, 3, 4$ [9].

$$\eta_1 = (X - Y)/\sqrt{2}, \quad \eta_2 = (X + Y - 2Z)/\sqrt{6}, \tag{1}$$

where $X = (x_1 + x_2 - x_3 - x_4)/2$, $Y = (y_1 - y_2 + y_3 - y_4)/2$ and $Z = (z_1 - z_2 - z_3 + z_4)/2$. Phase II is characterized by $\eta_1 = 0$, $\eta_2 \neq 0$ and phase III by $\eta_1 \neq 0$, $\eta_2 = 0$. The OP spans the E_u representation of the cubic point group O_h. The magnetisation **M** corresponds to the irreducible representation F_{1g}. Considering the symmetric products $[E_u \otimes E_u] = A_{1g} \oplus E_g$ and $[F_{1g} \otimes F_{1g}] = A_{1g} \oplus E_g \oplus F_{2g}$, we find that there are two independent constants describing biquadratic coupling between the

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OP and **M**. In an applied magnetic field **H**, the magnetic part of the free energy is

$$F_{\mathrm{M}} = A\mathbf{M}^2 - \mathbf{MH} + \gamma(\eta_1^2+\eta_2^2)\mathbf{M}^2 + \delta[(\eta_1^2-\eta_2^2)(2M_z^2 - M_x^2 - M_y^2) + 2\sqrt{3}\eta_1\eta_2(M_x^2 - M_y^2)]. \quad (2)$$

In all three phases, the product $\eta_1\eta_2$ is zero, and the inverse magnetic susceptibility is calculated to be

$$\chi_{zz}^{-1} = \chi_0^{-1} + 2\gamma(\eta_1^2+\eta_2^2) + 4\delta(\eta_1^2-\eta_2^2),$$
$$\chi_{xx}^{-1} = \chi_0^{-1} + 2\gamma(\eta_1^2+\eta_2^2) - 2\delta(\eta_1^2-\eta_2^2), \quad (3)$$

where $\chi_0 = (2A)^{-1}$ is the susceptibility of the high symmetry phase. In order to account for the experimentally observed behaviour at the two SPTs, we should assume $\delta \ll \gamma$ and $\gamma > 0$ [9]. Note the similarity between the present theory and the antiferromagnetic case, where the anomaly in the susceptibility also arises from biquadratic coupling between **M** and the antiferromagnetic vector.

The phenomenological model (2) is based on group-theoretical arguments only and does not distinguish between localized and itinerant electronic magnetism. We now restrict our attention to the itinerant case. The tight-binding approach describes the dispersion of electronic bands $\varepsilon_{\mathbf{k}}$ resulting from the electrostatic potential of the lattice. Overlap integrals, which constitute matrix elements of the tight-binding Hamiltonian, depend on the radius vector between neighbouring atoms [10]. Due to relations (1), this dependence can be expressed in terms of the OP, $\varepsilon_{\mathbf{k}}(\eta_1,\eta_2)$.

For simplicity, we consider a simple one-band model for small **k** around the center of the Brillouin zone. Group-theoretical considerations yield

$$\varepsilon_{\mathbf{k}}(\eta_1,\eta_2) = \hbar^2/2m^*[\mathbf{k}^2 + \alpha(\eta_1^2+\eta_2^2)\mathbf{k}^2] + \beta[(\eta_1^2-\eta_2^2)(2k_z^2 - k_x^2 - k_y^2) + 2\sqrt{3}\eta_1\eta_2(k_x^2 - k_y^2)]. \quad (4)$$

Here m^* is the effective mass in phase I, and α and β are constants which may be calculated by diagonalising the tight-binding Hamiltonian. Thus the effective mass components depend on the OP. In all three phases $\eta_1\eta_2 = 0$, therefore $m_x = m_y \neq m_z$.

The density of states at the Fermi level (including spin) is

$$D(E_{\mathrm{F}}) = \frac{1}{\hbar^2}\left(\frac{3m_x^2 m_z N}{\pi^4 V}\right)^{1/3}, \approx \frac{m^*}{\hbar^2}\left(\frac{3N}{\pi^4 V}\right)^{1/3}[1 - \alpha(\eta_1^2+\eta_2^2)], \quad (5)$$

where N/V is the number of conduction electrons per unit volume. Then the Pauli susceptibility at zero temperature is [11]

$$\chi_{\mathrm{P}} = \frac{g^2\mu_{\mathrm{B}}^2 m^*}{4\hbar^2}\left(\frac{3N}{\pi^4 V}\right)^{1/3}[1 - \alpha(\eta_1^2+\eta_2^2)]. \quad (6)$$

We note that up to the second order in the OP, only the isotropic part of the effective mass contributes to the density of states and Pauli susceptibility, but higher-order corrections will yield an anisotropic contribution from the OP. Moreover, spin–orbit coupling, according to band structure calculations [12,13], has a significant effect on the electronic spectrum, and also gives rise to anisotropy of the g factor [11]. Nevertheless, the simple model proposed here provides a mechanism of coupling between itinerant electrons and structural distortions, and the lowest order result (6) supports our assumption about the relative sizes of the phenomenological constants in Eq. (3).

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