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Tight-binding calculation of the magnetic moment of CrAs under pressure

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Abstract. We analyze the evolution of the local magnetic moment of the newly discovered pressure-induced superconductor CrAs, as a function of the applied pressure. Our theoretical method is based on a combination of the tight-binding approximation and the Löwdin down-folding procedure, which enables us to derive a low-energy effective Hamiltonian projected onto the Cr-subsector. We set up our calculations by considering several sets of *ab initio* derived hopping parameters, corresponding to different volumes of the unit cell, and use them to obtain the simulated pressure-dependence of the Cr magnetic moment, which is evaluated within a mean-field treatment of the Coulomb repulsion between the electrons at the Cr sites. Our calculations show good agreement with available experimental data, both for the normal phase measured 1.7 μ_B for Cr magnetic moment, and concerning the observed reduction of its amplitude for values that exceed the characteristic critical pressure.

1. Introduction

Recently, superconductivity was discovered in the Cr-based compound CrAs under pressure [1, 2]. Similarly to other superconducting materials, such as heavy-fermion compounds [3], high transition-temperature cuprate superconductors [4], strontium ruthenate superconductors [5] and iron-pnictide superconductors [6], the superconductivity in the CrAs appears near a magnetic quantum critical point. Wu *et al.* reported the appearance of superconductivity at $T_c \sim 2$ K for a critical pressure $P_c \sim 8$ Kbar, when the antiferromagnetic transition, that sits at $T_N \sim 265$ K under ambient pressure, is totally suppressed [1, 7]. According to Kotegawa *et al.*, the superconducting transition temperature has its maximum at about 1 GPa, and decreases at higher pressure, showing a well-known dome-shaped phase diagram [1, 2, 8, 9, 10, 11, 12, 13, 14, 15]. Furthermore, nuclear quadrupole resonance under pressure studies indicate a slow decrease, with a large value close to P_c, of the internal field in the magnetic state when the pressure increases. This indicates that the pressure-induced suppression of the magnetic order is a first order phase transition [8].

All these properties point towards a possible unconventional mechanism of formation of Cooper pairs, likely due to the interaction with magnetic fluctuations in the material near

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the quantum critical point [1]. However, measurements of muon spin rotation on CrAs powder samples reveal that there is a coexistence region, in the intermediate pressure regime, where the magnetic and the superconducting phases are well separated, and this, together with the observation that the superfluid density ρ_s scales with the critical temperature as $\sim T_c^{3.2}$, seems to bring towards a conventional mechanism of pairing in CrAs [16].

Neutron diffraction measurements [17, 18] establish that CrAs is a non collinear helimagnet, with a substantial Cr moment of 1.7 μ_B at ambient pressure that lies essentially within the *ab* plane, the propagation vector being directed along the *c* axis [18]. The magnetic structure is formed by four spirals along the *c* axis, one for every Cr-atom in the primitive cell, with a well-defined phase angle between the spirals. Measurements of the magnetic properties versus the pressure indicate that the average ordered magnetic moment decreases from 1.7 μ_B at ambient pressure to 0.4 μ_B at the critical pressure $P_c \sim 0.7$ GPa, where the magnetic order is totally suppressed [18]. Furthermore, Shen *et al.* showed that CrAs exhibits a spin reorientation in addition to an abrupt decrease of the magnetic propagation vector at the critical pressure. At this pressure the system moves from a configuration where the magnetic moments lie in the *ab* plane to one where they lie in the *ac* plane. This magnetic phase transition coincides with the emergence of bulk superconductivity. At higher pressure, CrAs approaches to the optimal superconductivity regime, where the magnetic moments tend to be aligned antiparallel between nearest neighbor ions [17].

Moreover, the helimagnetic order is correlated to structural and electronic degrees of freedom [17]. At $T_N \sim 265$ K, in addition to the magnetic transition, there are discontinuous changes of the lattice parameters [1], suggesting that electronic, magnetic and structural properties are correlated as in many transition metal compounds [19, 20, 21, 22].

As far as the theoretical results are concerned, *ab initio* calculations based on first-principles density functional theory strengthen the experimental trend that the magnetic and electronic properties strongly depend on the size of the unit cell [23]. Furthermore, the reasonable agreement for the magnetic moment obtained within this approach and the fact that the electric resistivity of CrAs doesn't seem to exceed the Mott-Ioffe-Regel limit [24] suggest that the CrAs could be considered as a weakly correlated material [23]. Indeed the Cr atoms in the CrAs compound can be considered closer to the metallic Cr than the strongly correlated ionic Cr compounds [25].

Besides, a method that combines the tight-binding approximation and the Löwdin down-folding procedure [26, 27] has been adopted to study some properties of CrAs [28]. Within this approach, the parameters entering the tight-binding matrix are the *ab initio* derived overlap integrals for orbitals, whereas the Löwdin technique allows us to integrate out all low-lying arsenic degrees of freedom and to get an Hamiltonian whose rank is definitely lower than that of the starting tight-binding matrix.

Taking advantage from this procedure, here we will focus on the magnetic properties of CrAs, investigating the behaviour of the Cr magnetic moment as a function of the variations of the volume of the primitive cell, as induced by applied external pressure. The paper is organized as follows: in the next section we will introduce the model Hamiltonian adopted and we will introduce the Löwdin down-folding procedure; Sec. III will be devoted to a discussion of the obtained results and to some remarks.

2. Model Hamiltonian and Löwdin down-folding procedure

CrAs exhibits either an hexagonal NiAs-type (B81) structure or an orthorhombic MnP-type (B31) structure. At ambient pressure, the system presents an orthorhombic MnP-type (B31) structure, and the unit-cell lattice parameters are a=5.649 Å, b=3.463 Å and c=6.2084 Å [1]. The primitive cell of the CrAs is reported in Fig. 1, and it consists of four Cr atoms and four As atoms. To study the magnetic properties of the material, and in particular the trend of the

magnetization as a function of the external pressure, we start from the following tight-binding Hamiltonian:

$$H = \sum_{i,\sigma} \epsilon_i c^{\dagger}_{i\sigma} c_{i\sigma} - \sum_{i,j,\sigma} t_{ij} (c^{\dagger}_{i\sigma} c_{j\sigma} + h.c.) + U \sum_{i} n_{i\uparrow} n_{i\downarrow} \,. \tag{1}$$

This Hamiltonian is the sum of three contributions: the first one describes the on-site term, ϵ_i being the energy at each Cr or As lattice site; the second one takes into account the hopping processes of the electrons with spin σ between the i and j sites of the Cr or As ions; finally the last term describes the Coulomb repulsion between electrons on the same Cr site having opposite spin.

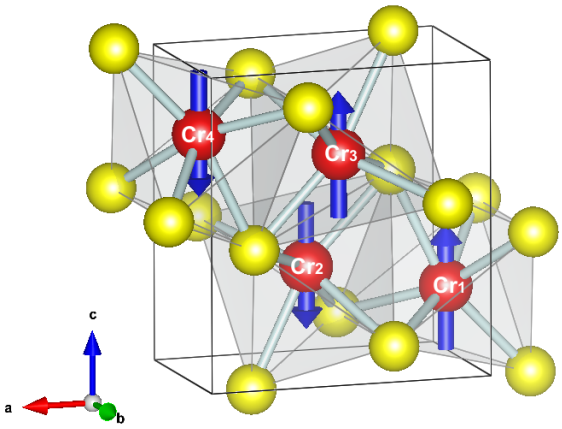


Figure 1. Primitive cell of the CrAs at ambient pressure. Cr and As atoms are shown as red and yellow spheres, respectively, while the arrows represent the spins of the Cr atoms in the magnetic ground state and in the collinear approximation.

Since the primitive cell contains four Cr atoms and four As atoms, and due to the fact that the orbitals entering the general Hamiltonian in Eq.(1) are the d orbitals for Cr atoms and the p orbitals for As atoms, the corresponding tight-binding matrix Hamiltonian is given by a 32×32 matrix. Nevertheless, since the As bands are located above and below 2 eV from the Fermi level [23], we use the Löwdin down-folding procedure to get the energy dispersion of the Cr d bands only. Within this approach, the whole Hamiltonian is partitioned considering a 20×20 matrix that describes the d-d hoppings among Cr ions, a 12×12 one describing the p-p As ions hoppings, and two sub-matrices that correspond to the d-p hoppings from Cr to As ions and vice-versa. The energy spectrum of the $3d$ bands can be then obtained by integrating out all low-lying As $4p$ degrees of freedom and diagonalizing the renormalized 20×20 Hamiltonian projected onto the Cr orbitals.

We point out that we have considered the nearest neighbour hoppings, the second nearest neighbour hoppings along the x-direction and the diagonal part of the second nearest neighbour hoppings along the y and z direction, confining the analysis to the parameters whose values range from 80 meV to 1 eV.

Since CrAs may be considered as a weakly correlated material, we can treat the Coulomb term in the mean-field approximation. Thus, the Hamiltonian of Eq.(1) becomes

$$H = -\sum_{i,j,\sigma} \tilde{t}_{ij} (\tilde{c}^{\dagger}_{i\sigma} \tilde{c}_{j\sigma} + h.c.) + U \sum_{i} (\tilde{n}_{i\uparrow} \langle \tilde{n}_{i\downarrow} \rangle + \tilde{n}_{i\downarrow} \langle \tilde{n}_{i\uparrow} \rangle - \langle \tilde{n}_{i\uparrow} \rangle \langle \tilde{n}_{i\downarrow} \rangle) \,, \tag{2}$$

where $\tilde{t}_{ij}$ are the renormalized hopping amplitudes obtained after the application of the Löwdin procedure, $\tilde{c}^{\dagger}_{i\sigma}$ and $\tilde{c}_{i\sigma}$ refer to the creation and annihilation operators of renormalized d Cr

electrons, $\langle \tilde{n}_{i\uparrow} \rangle$ and $\langle \tilde{n}_{i\downarrow} \rangle$ are the average, in the ground state, of the electron number operator for spin up and down at the i lattice site, respectively.
To obtain the magnetization in the G-type configuration emerging from *ab initio* calculations [23], we performed a self-consistent procedure according to the following scheme: we assign initial conditions for $\langle \tilde{n}_{i\uparrow} \rangle$ and $\langle \tilde{n}_{i\downarrow} \rangle$, and use them to evaluate improved expectation values, this procedure running until convergence with the requested accuracy is achieved.

3. Results and discussion

To simulate the effect of the external pressure acting on the system, we have performed our calculations at different values of the lattice constants, corresponding to different values of the volume of the primitive cell. This procedure is corroborated by the experimental data in Ref. [17] where it is reported the magnetic moment of the CrAs as a function of the pressure as well as of the lattice constants. From these data we have been able to plot in Fig. 2 the experimental pressure dependence of the volume of the primitive cell. We use these experimental results as input for our calculation.

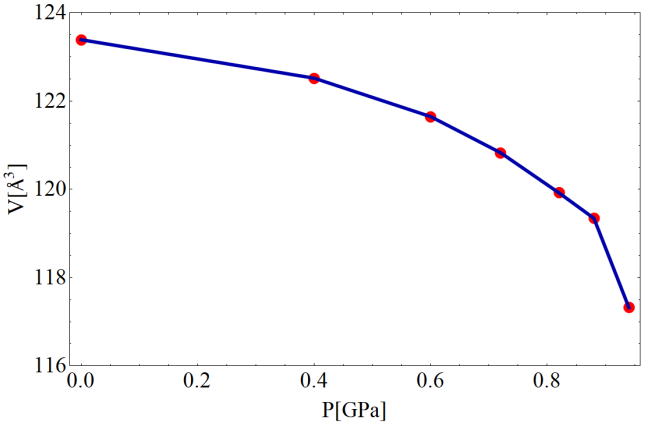


Figure 2. Volume of the primitive cell of the CrAs as a function of the external pressure acting on the system. The values are those reported in Ref. [17].

Within our scheme, to obtain the magnetic moment as a function of the external pressure, we choose the corresponding lattice parameters and perform the above mentioned self-consistent method. According to *ab initio* calculations, the value of the Coulomb interaction has been fixed at U=0.37 eV. The results of our simulations are reported in Fig. 3, together with the available experimental data [17] and the outcomes of the *ab initio* calculation [23].

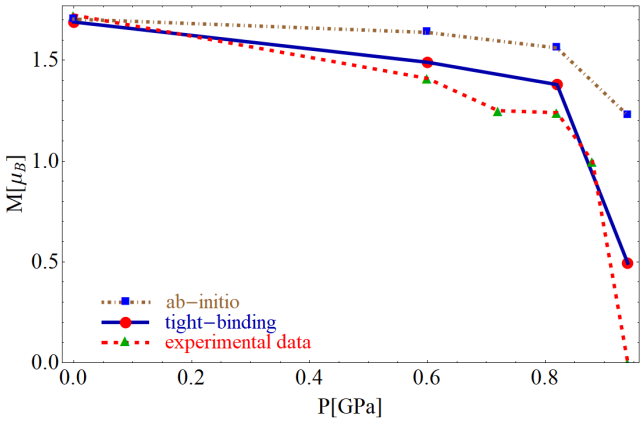


Figure 3. Magnetization of CrAs as a function of the external pressure. The blue line indicates the theoretical results, the brown dot-dashed line comes from the *ab initio* simulation [23], while the red dashed line is obtained from the experimental data in Ref. [17].

From this figure we notice that a general overall agreement with the experimental data is obtained as far as it concerns both the amplitude of the Cr magnetic moment and its reduction

upon applied pressure. According to the results of Refs. [23, 28], we infer that the magnetic moment reduction could be linked to the increase of the Cr-bandwidth. To clarify this point, we have plotted in Fig. 4 the Cr-bandwidth W as a function of the pressure for some values of the Coulomb repulsion, namely U=0.35, 0.37 and 0.39 eV. There are two trends that can be distinguished in Fig. 4. Below P=0.8 GPa the bandwidth smoothly decreases with increasing P, with a maximum percentage variation of 3% at 0.6 GPa. In correspondence, the evolution of the magnetization, as shown in Fig. 3, is almost constant. For external pressures that exceed P=0.8 GPa, the bandwidth displays a sudden increase corresponding to the dramatic reduction of the local magnetic moment, for all the values of U investigated. We also notice that the minimum of the Cr-bandwidth at P=0.6 GPa coincides with the maximum strength of the antiferromagnetic order [23]. Nevertheless, we mention that other quantities may control the magnetic properties of CrAs, such as the bandwidth of the single bands crossing the Fermi level or the spin-orbit coupling. For completeness, we notice that in Refs. [7, 17] it is mentioned that the magnetic moments of the Cr atoms with shorter distance are antiparallel, indicating a strong antiferromagnetic coupling between these Cr atoms, in agreement with the results here presented. We finally point out that the local magnetic moment, evaluated via the mean-field treatment of the Coulomb repulsion within tight binding method, shows even a better agreement with the experiments, compared with the LDA calculations, as shown in Fig. 3 [23].

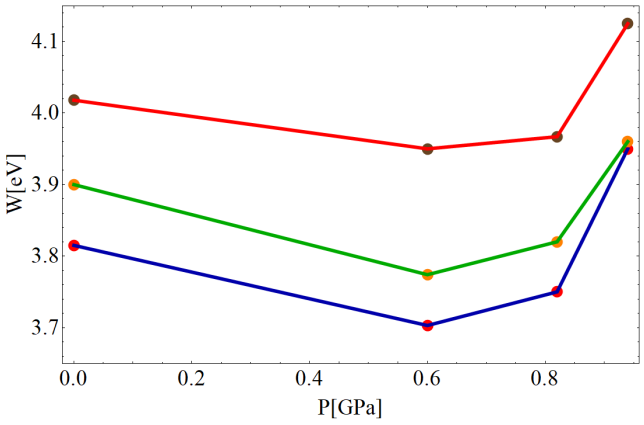


Figure 4. Bandwidth W of CrAs as function of the pressure for U=0.35 eV (blue line), 0.37 eV (green line) and 0.39 eV (red line).

It is worth stressing that even if *ab initio* calculations are available, they are not delivered in a form useful as the single-particle term of a model Hamiltonian that describes the low-energy excitations, whereas obtaining an effective Cr-bands only model is highly non-trivial [29]. Here, we have neglected the nontrivial details of the LDA band structure and have used the simplest possible tight-binding model with hopping integrals between only nearest and next-nearest neighbors. Hence, within the Löwdin procedure we have reduced the full tight-binding for As and Cr ions to Cr d bands only, deriving manageable expressions for d-band dispersions near the Fermi level. This result turns out to be readily used in calculating physical quantities depending on the explicit form of the energy spectrum, as for instance the in-plane resistivity [28]. Furthermore, we are confident that this approach could be used as the single-particle term in more sophisticated Hamiltonians containing for instance Coulomb correlations, such as the Hund coupling, as well as to search for the superconducting instability within the standard broken-symmetry Hartree-Fock scheme [30, 31].

Finally, the overall agreement between the electric resistivity obtained using the procedure before specified and the experimental data as well as the Cr magnetic moment here discussed suggests that this material is a weakly correlated compound whose magnetic and electric properties are suitably described by a tight-binding approach. Nevertheless, to get the helimagnetic configuration exhibited by CrAs the As spin-orbit coupling has to be included.

Moreover, the introduction of the As spin-orbit coupling can generate an effective Dzyaloshinskii Moriya interaction between the Cr atoms [32]. Calculations in this direction are in progress and will be presented elsewhere.

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