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# Self-consistent electronic structure of CeCoIn<sub>5</sub> considering Coulomb correlation effects

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## Abstract

The electronic structure of CeCoIn<sub>5</sub>, derived from a self-consistent calculation by means of a local density approximation (LDA) potential within the augmented plane wave framework, is used as an initial state. Considering this state, a mean field approximation from the multiband Hubbard Hamiltonian has been considered in order to include the strong correlation effects and a new self-consistent band structure has been determined. The results yield a spectrum where the splitting of the Hubbard bands is clearly displayed and the two-dimensional character of the electronic structure of this compound is obtained in a better way with respect to the structure attained with the simple LDA calculation.

## 1. Introduction

CeCoIn<sub>5</sub> has been discovered to be a superconductor [1–4], whose transition temperature ( $T_c = 2.3$  K) at room pressure is the highest of the superconducting heavy-fermion compounds. This Ce compound crystallizes into the HoCoGa<sub>5</sub> tetragonal crystal structure [1–4]. The lattice parameters are  $a = 4.62$  Å and  $c = 7.56$  Å and the experimental data available seem to indicate that the electronic dynamic occurs fundamentally in a two-dimensional way where alternating layers [1–4] of CeIn and CoIn<sub>4</sub> seem to be almost independent, giving this compound a characteristic 2D structure similar to that of the LaCuO<sub>4</sub> systems. The unit cell of this compound contains a molecule where the Ce atoms are located at the vertexes, forming a rectangle with an In atom in its centre. The electronic structure from the local density approximation (LDA) shows that the electrically active electrons close to  $E_F$  belong to this CeIn rectangle. Another group, CoIn<sub>4</sub>, is located between the two layers of CeIn and seems to be more responsible for the magnetic properties than the CeIn layer. The high electronic specific heat in the normal state above the transition temperature, i.e.  $T > 2.5$  K, is  $290 \text{ mJ mol}^{-1} \text{ K}^{-2}$ , indicating its heavy-fermion behaviour [1–4]. In addition, this specific

heat for temperatures less than  $T_c$ ,  $C/T = 0.04 + 0.25T \text{ J mol}^{-1} \text{ K}^{-2}$ , is a clear feature of unconventional superconductivity, which can imply node lines in the superconducting gap, possibly with anisotropic pair potential arising from magnetic interactions.

The first step in determining the electronic structure is the LDA Hamiltonian [5, 6]. Let us summarize the characteristics of the LDA electronic structure in this way: the band-structure from the local density formalism [5, 6] presents a width of around 8.2 eV from the band bottom up to  $E_F$ . The conduction band has 12 extended subbands that fundamentally arise from the cobalt d orbitals, and s and p orbitals from the In atoms. These extended bands are strongly hybridized and the Fermi level is at 0.44 Ryd with respect to the zero of the muffin-tin potential. At around  $E_F$  seven Ce f bands appear. The f bands cross the Fermi level as well as the d bands of cerium and cobalt, and the s and p bands of indium. This implies slightly different occupations for the corresponding bands within each  $l$ -angular momentum orbital because the crystal symmetry slightly breaks the  $l$ -degeneracy, although the band dispersion effects soften the splitting. In the case of the f orbitals, the spatial localization of the charge implies that small variations in the gravity centres along with different hybridizations produce some differences in their occupations. The narrowness of the flat f bands is clear [5] and this circumstance implies that the inclusion of the Hubbard Hamiltonian is legitimate. The d band states coming from the Co atom are below the  $E_F$  and present an intermediate electronic correlation with sufficiently localized states that are, however, obviously less than the f states. This implies that the Coulomb interaction in the same atom, the familiar  $U$ , is also large although clearly less than that of the f states. As a consequence, in this calculation we have not included a Hubbard pseudopotential for these d bands. The localization of the 4f states and their partial orbital occupation have as a consequence that the crystal field effects are smaller than the well known  $U$ -energy, which for the Ce compounds becomes around 7 eV. Therefore, the second step for the analysis of this electronic structure of CeCoIn<sub>5</sub> is the determination of the Hubbard splitting in lower and upper bands, which constitutes one of the most significant strong correlation effects within the electronic structure of these compounds. The calculation of these effects and the analysis of the resulting electronic structure is the main goal of this paper.

## 2. Band structure with strong correlation pseudopotential

At the present time, to the authors' knowledge, only LDA calculations [5, 6] have been realized in this intermetallic alloy and there is no calculation in the literature for this compound within this LDA +  $U$  approximation. Therefore, this could be a new step for understanding the electronic structure of the normal state of this heavy-fermion superconductor. The main reasons provided by the experimental data for spending time on the band structure calculations including strong correlation effects are that

- (i) the f states of Ce are located at  $E_F$  and this situation is a clear scenario, present in many different materials, where the Coulomb correlation effects are substantial in the normal and superconductor states,
- (ii) the simple LDA is not able to give the upper and lower bands which will foreseeably appear in direct and inverse photoemission spectroscopies and
- (iii) another important failure of the LDA calculation is that because the LDA Hamiltonian does not split the different f symmetries and as these states are dominant in  $E_F$ , the planar structure of this compound is not clearly shown in the LDA calculation and can only be seen as a tendency toward this two-dimensional dynamic.

The Coulomb correlation effects imply that the occupation differences in the  $f$  orbitals can induce sensible variations in the corresponding Coulomb correlation pseudopotentials [7, 8]. The existence of different pseudopotentials for each  $f$  symmetry implies that the procedure for calculating the band structure has to be self-consistent, since these pseudopotentials can produce drastic variations in the energy position of the corresponding  $f$  orbitals. On the other hand, the LDA potential,  $V_{\text{LDA}}$ , is also strongly dependent on the occupation of the  $f$  level, and generally the actions of this potential are in competitive directions to the Hubbard pseudopotentials. As is well known, the Coulomb correlation effects should be included in the electronic structure starting from a ground state electronic structure where the Hubbard Hamiltonian effects should be excluded. The pseudopotential [7, 8] for each  $f_j$  orbital is  $V_j = U(n_f - n_j)|f_j\rangle\langle f_j|$ , where  $|f_j\rangle\langle f_j|$  is the projector for the  $f$  orbital with a given symmetry.  $U$  is the Coulomb Hubbard energy;  $n_j$  is the self-consistent obtained occupation of the  $f_j$  orbital (the seven  $f_j$  orbitals should be compatible with the crystal symmetry group), and  $n_f = \sum_j n_j$  is the total occupation within a given atom of the  $f$  level. The self-consistency, considering the two potentials  $V_{\text{LDA}}$  and  $V_U = \sum_j V_j$ , has many variants and the methodology followed in this paper consists of a calculation of  $V_U$  pseudopotential and  $V_{\text{LDA}}$  for each iteration. We initialize the LDA +  $U$  calculations starting from the last iteration LDA potential. The solution of the corresponding Schrödinger equation, considering the two potentials, is realized within a modified augmented plane wave procedure [8]. As stated above, one of the main objectives of this calculation is to obtain the Hubbard splitting that yields the characteristic lower and upper Hubbard bands. The action of the  $V_U$  potential in each iteration is to locate each  $f_j$  orbital as an independent  $f_j$  level with a different energy, which is constituted as a  $f_j$  band gravity centre ( $\Gamma_j$ ). So, a larger previous occupation leads to a smaller energy increase of the corresponding  $f_j$  level due to  $V_U$ . In contrast, the LDA potential does not distinguish the different  $f_j$  symmetries and therefore yields a degenerate  $f_j$  level with some small splitting of the corresponding band centres due to the different hybridization and the different effects of the crystalline field in each  $f_j$  symmetry.

The two competing and contradictory actions of  $V_U$  and  $V_{\text{LDA}}$  in each iteration require a subtle and complex strategy in the self-consistent calculation. It is well known that in the cases in which the Coulomb correlation is larger than the crystal field effects several convergent solutions exist that are obtained self-consistently. In all self-consistent processes, the total charge density for obtaining the corresponding potential for a given  $i$  iteration has to be constructed by means of the golden rule:  $\rho(\vec{r}) = \alpha\rho^i(\vec{r}) + (1 - \alpha)\rho^{i-1}(\vec{r})$ , where  $\rho^i(\vec{r})$  and  $\rho^{i-1}(\vec{r})$  are the charge densities obtained in the iterations,  $i$  and  $i - 1$ , respectively, and the  $\alpha$  parameter, which can take any value larger than zero and less than unity, does not modify the final result although it can modify the difficulties for obtaining it. In the case of two competing potentials such as  $V_U$  and  $V_{\text{LDA}}$ , the occupations of the  $f_j$  orbitals in the final results can depend on the  $f_j$  occupations in the initial state (i.e. the final results of the LDA calculation) and the  $\alpha$  parameter. Consequently, it is possible to arrive at different final self-consistent solutions according to the chosen  $\alpha$  parameter in the LDA +  $U$  calculations. Even for small differences in the value of this  $\alpha$  parameter, the distribution of  $f_j$  occupations is different, although the final result of  $n_f = \sum_j n_j$  only suffers slight modifications for the different values of  $\alpha$ . Finding the optimal initial  $f_j$  occupations and the value for the  $\alpha$  parameter is calculation work, maybe without physical significance in itself, but strictly necessary in order to legitimate the final result. The optimal results for the calculation process should be when the final distribution of the  $f_j$ -occupations is such that the total energy of the corresponding self-consistently electronic structure is minimal. Therefore, it is important to determine the total energy of the electronic structure as a function of those physical variables that can be susceptible to being modified in each self-consistent procedure.

### 2.1. Total energy

The total number of conduction electrons,  $n_T$ , is

$$n_T = n_f^0 + n_f^- + \int_b^{E_F} N_{\text{spd}}(\epsilon) d\epsilon, \quad (1)$$

where  $n_f^0$  is the electron number in partially occupied  $f_j$  orbitals,  $n_f^0 \equiv \sum_{j=1}^{\mu} n_j$ , with  $n_j = \int_b^{E_F} N_j(\epsilon) d\epsilon$  being the occupation of a given  $f_j$  symmetry,  $N_j(\epsilon)$  the  $f$  DOS of this  $f_j$  orbital, and  $b$  the conduction band bottom;  $n_f^-$  is the electron number in totally occupied  $f_j$  orbitals [ $n_f^- = \sum_{j=\mu+1}^{\mu+m} n_j \simeq m$ ];  $N_{\text{spd}}(\epsilon)$  is the density of states excluding all the  $f_j$  states. Therefore, the number of  $f$  electrons is  $n_f = n_f^0 + n_f^-$ . The Fermi level  $E_F$  can vary for each self-consistent process, and the charge conservation implies that  $D_F \delta E_F + \delta n_f^0 \simeq 0$  where  $D_F \equiv N_{\text{spd}}(E_F)$  is the DOS of non- $f$  orbitals in the Fermi level.

Each LDA +  $U$  self-consistent process can start with arbitrary  $n_j$  occupation numbers. One can choose those  $n_j$  occupations coming from the LDA calculations. With these initial  $n_j$  occupation numbers and using a modified APW method [8], the energy bands, the Fermi level and new values of  $n_j$  are calculated, and a new iteration can then start. Each  $f_j$  orbital has two gravity band centres, with and without considering the potential of the other  $f$  electrons. These two band centres are called  $\Gamma_j$  and  $\gamma_j$ , respectively, and their relationship is  $\Gamma_j = \gamma_j + n_f U_0 + U(n_f - n_j)$ , where  $U_0$  is given approximately by the  $F^0$  Slater integral and  $U(n_f - n_j)$  is the first order self-energy correction within the Hubbard Hamiltonian for each  $j$  symmetry of the  $f$  orbitals.

In each self-consistent process, the total energy is given by

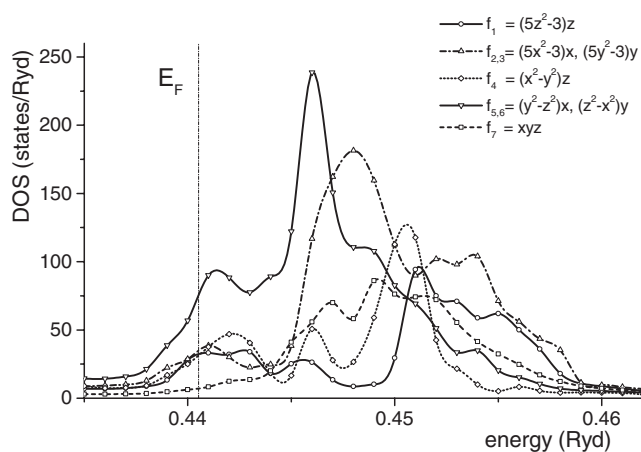
$$E_T = \int_b^{E_F} \epsilon N_{\text{spd}}(\epsilon) d\epsilon + \sum_{j=1}^{\mu} \int_b^{E_F} \epsilon N_j(\epsilon) d\epsilon + \sum_{j=\mu+1}^{\mu+m} \Gamma_j. \quad (2)$$

Consequently, it is clear that the valid results should arise from a self-consistent process that ensures the minimal total energy in equation (2). In this paper we give the definite results, i.e. the electronic structure whose total energy from equation (2) is minimal when the convergence in the self-consistent process is obtained.

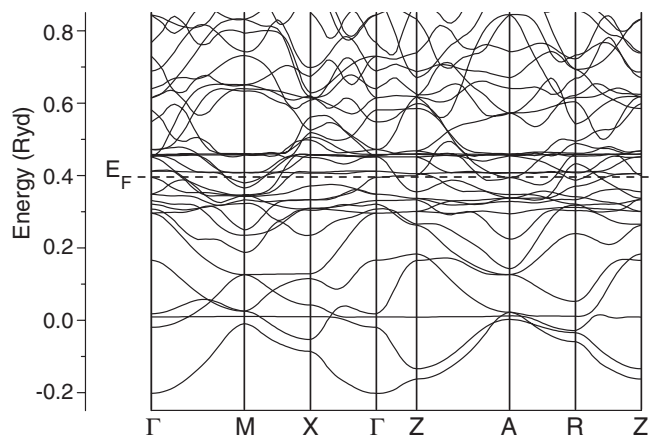
## 3. Results

Figure 1 shows the partial  $f$  DOS distributed by the different  $f_j$  symmetries obtained from the LDA calculation. In this crystal symmetry group the compatible  $f_j$  symmetries are  $f_1 = 5z^3 - 3z$ ,  $(f_2, f_3) = (5x^3 - 3x, 5y^3 - 3y)$ ,  $f_4 = x^2z - y^2z$ ,  $(f_5, f_6) = (y^2x - z^2x, z^2y - x^2y)$ , and  $f_7 = xyz$ . According to these results, the occupations of the  $f$  orbitals,  $f_1$ ,  $(f_2, f_3)$ ,  $f_4$ ,  $(f_5, f_6)$ , and  $f_7$ , are 0.16, (0.12, 0.12), 0.23, (0.17, 0.17), and 0.08, respectively. Each of these 4 $f$  states tends to be hybridized with those orbitals of the In(1), which are located in the same CeIn layer, although these states are also hybridized with the states of the other In atoms. This LDA band structure is taken into account for considering the initial conditions for the LDA +  $U$  calculation.

In figure 2, we give the band structure deduced from the two potentials,  $V_{\text{LDA}}$  and  $V_U$ , described above and corresponding to the electronic structure of minimal total energy. In comparison with figure 1 of our previous paper [5], one can see that, in addition to the 12 extended bands that appear in the LDA calculation, in figure 2 the corresponding band to the  $f_4$  orbital  $(x^2 - y^2)z$  appears 0.4 Ryd below the  $E_F$ , weakly hybridized with the  $s$  and  $p$  orbitals of In and practically without hybridization with the  $d$  states of Co. The dominance of the  $f$  states in energies close to, but larger than,  $E_F$  is clearly shown in figures 3 and 4. In figure 3,



**Figure 1.** Partial density of states for each  $f_j$  orbital from the LDA calculation.



**Figure 2.** LDA +  $U$  band structure of CeCoIn<sub>5</sub>.

we present the total DOS corresponding to the band structure of figure 2. The differences with the corresponding LDA results of [5] are clear:

- (i) the  $f_4 = (x^2 - y^2)z$  peak located 0.4 Ryd below  $E_F$  does not appear in the LDA calculation,
- (ii) the splitting of the unoccupied  $f$  peaks, and the location of  $E_F$  just below the peak corresponding to the two-dimensional  $f$  subspace whose symmetry is  $(f_2, f_3) = (5x^3 - 3x, 5y^3 - 3y)$ , and
- (iii) the totally unoccupied  $f$  states are split from those arising from the  $(x^2 - y^2)z$  states with an energy of 0.45 Ryd, which is the  $U$ -value considered in the construction of the  $V_U$  pseudopotential in all our calculations.

The fact that there is no relevant hybridization of the cobalt  $d$  orbitals with the cerium  $f$  band states, and also that the electrical dynamic is governed by the  $(5x^3 - 3x, 5y^3 - 3y)$  states which are the closest states to the  $E_F$ , allows us to think of this material as a double-layered compound with two almost independent structures; this idea is in accordance with some experimental conclusions [1–4].

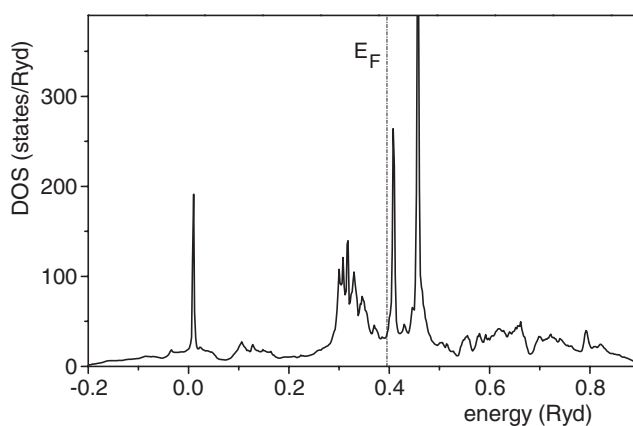


Figure 3. LDA +  $U$  total density of states.

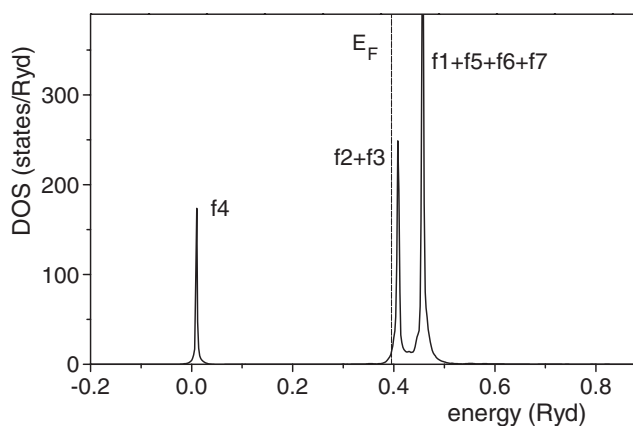
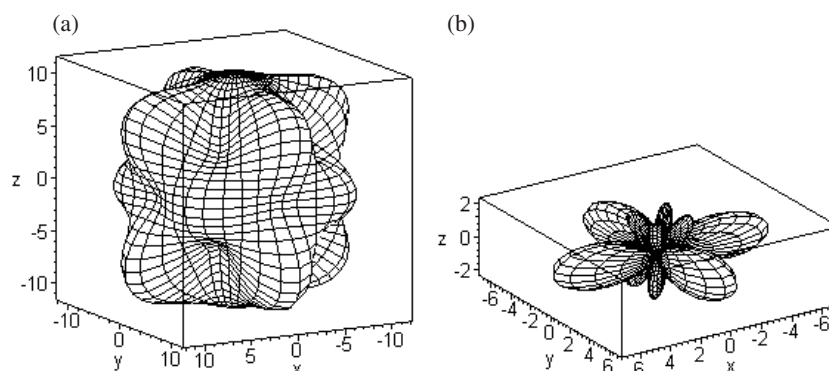


Figure 4. LDA +  $U$  partial density of the states for each  $f_j$  orbital.

In figure 5, we give a polar representation of the directional dependence of the charge density in the LDA and LDA +  $U$  calculations. We represent the charge density  $\rho(\hat{\mathbf{r}}) = \sum_{\mathbf{k}} \sum_j |\langle \Psi_{\mathbf{k}}(\hat{\mathbf{r}}) | f_j(\hat{\mathbf{r}}) \rangle|^2$ , where  $\Psi_{\mathbf{k}}(\hat{\mathbf{r}})$  is the angular part (inside the muffin-tin spheres) of the self-consistent solution of the Schrodinger equation in the solid and  $f_j(\hat{\mathbf{r}})$  are the angular parts of the  $f_j$  symmetries. The  $\mathbf{k}$ -index runs over all the occupied states in the first Brillouin zone. In figure 5(a) we draw the directional dependence of the charge density of the LDA calculation. This charge density of figure 5(a) does not display any two-dimensional nature of the charge distribution. In contrast, in figure 5(b), the tendency to bidimensionality in the  $x$ - $y$  plane of the charge density distribution is clearly shown.

The facts described in the above item (ii) concerning our results in figures 3 and 4 and those calculations shown in figure 5 are a quantitative theoretical ratification of the two-dimensionality of this material. This point is a clear improvement with respect to the LDA result concerning the dimensionality of this compound (the LDA Hamiltonian is not able to determine the true directionality of the charge density and the vectorial overlapping of the states close to  $E_F$  [5]).



**Figure 5.** Polar representation of the directional dependence of the charge density corresponding to the band structures: (a) LDA and (b) LDA +  $U$ .

#### 4. Final comments

Equation (2) is obtained taking into account a first perturbation order in the evaluation of the total energy with the Hubbard Hamiltonian. In other higher orders the expression of  $E_T$  only differs in the last term,  $\sum_{i=\mu+1}^{\mu+m} \Gamma_i$ . However, as is well known, the mean field can yield sufficiently valid results for the total energy of the systems. The inclusion of higher orders can be important in order to obtain self-energy effects, which can be responsible for a part of the mass renormalization of the states close to  $E_F$ . This point could be another step of the process of achieving the electronic structure of this material. Nevertheless, the heavy-fermion properties with the superconducting behaviour seems to be concatenated in these types of materials. This induces us to think that the mass renormalization is not only due to a self-energy effect in the electronic structure near  $E_F$ , but cooperative effects arising from spin fluctuations can play an important role in both the thermodynamic properties and the fermionic coupling mechanism. In the double structure, the CoIn<sub>5</sub> planes present a 2D lattice in which a spin field can arise from the d states of Co and the extended bands are formed by the s and p orbital band states of In(2), located in the centre of this square spin lattice. The dynamics of this CoIn<sub>4</sub> layer have the ingredients of a Kondo lattice systems in two dimensions, which can be treated by means of a double-exchange Hamiltonian,  $J_{\text{Kondo}}$  plus the induced  $J_{\text{RKKY}}$ . The existence of these CoIn<sub>4</sub> layers may imply the combination and even the competition with another physically more complex substructure arising from the CeIn planes. As is well known, the double-exchange materials present strong instabilities and different quantum phases [9] that appear due to small variations in the dynamic parameters governing the spectra of this general Hamiltonian. The inter-relationship of these phases corresponding to each substructure depends on the symmetry of the  $f_j$  states near  $E_F$  as well as their hybridization with other states. These facts are relevant to the nature of the band structure, and allow the Hamiltonian parameters of the Kondo/Heisenberg system to be quantitatively obtained, which may provide a mean by which to analyse this material's unconventional superconductivity, and whose treatment and study will be set out from this mixed exchange model in a future study.

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