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J. Phys.: Condens. Matter **10** (1998) 11565–11570. Printed in the UK PII: S0953-8984(98)96565-1

The Fermi surface of 5f-electron compounds

T Maehira†, M Higuchi‡, M Nakamura§ and A Hasegawa‖

† Graduate School of Science and Technology, Niigata University, Niigata, 950-2181, Japan
‡ Department of Physics, Tohoku University, Sendai, 980-8576, Japan
§ PNC, Tokai, Ibaraki, 319-1194, Japan
‖ Faculty of Science, Niigata University, Niigata, 950-2181, Japan

Received 1 June 1998

Abstract. Energy band-structure calculations for UC are improved by determining the lattice constant and the U and C APW sphere radii so as to minimize the total energy. In the energy band structure so derived, the Fermi surface consists of hole sheets centred at the X and L points and electron sheets centred at the W points. The hole sheets centred at the X points and the electron sheets centred at the W points can explain the experimental result for the de Haas–van Alphen effect qualitatively. Quantitative comparison reveals, however, that the hole and electron sheets are too large in size. This result implies that a careful treatment of the exchange and correlation interaction between electrons beyond the local-density approximation may be necessary.

1. Introduction

NaCl-type uranium carbide UC as well as many other NaCl-type actinide compounds have attracted interest because of their unusual physical properties such as a high melting point, hardness, brittleness and metallic conduction [1]. Unlike many other NaCl-type uranium compounds, UC is paramagnetic. The low-temperature electronic specific heat coefficient γ is moderately large, i.e., $\gamma = 20$ mJ K^{-2} mol^{-1} [1]. Owing to the simple crystal structure and paramagnetism, UC is appropriate as the subject for a fundamental study of the nature of the 5f electrons in actinides.

By using a self-consistent relativistic augmented-plane-wave (APW) method and the exchange and correlation potential in the local-density approximation, it is predicted that UC may be a semimetal with the Fermi surface consisting of three hole pockets in the C 2p valence band which are centred at the X points in the Brillouin zone and six electron pockets in the U 5f conduction band which are centred at the W points [2]. These hole and electron sheets of the Fermi surface can explain reasonably well the experimental result of an angular dependence of the frequencies in the de Haas–van Alphen (dHvA) effect [3].

The previous band calculation was too simple, however, because it was carried out for the experimental lattice constant only. Whether the total energy was minimum or not was not investigated. In this paper, we improve on the band calculation for UC by determining the lattice constant as well as the U and C APW sphere radii so as to minimize the total energy within the framework of the local-density approximation.

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2. The method of band calculation

In this calculation, a relativistic symmetrized APW method is used with the muffin-tin approximation to the spatial from of the potential and the electron density [4]. The exchange and correlation interactions are treated within the density functional theory in a local-density approximation [5]. This method has been derived by improving on the Loucks relativistic band theory [6] in terms of symmetrization of the bispinor-type basis functions with double-multiplier space groups. For the application of this method to many rare-earth compounds, the reader is referred to the review by Onuki and Hasegawa [7].

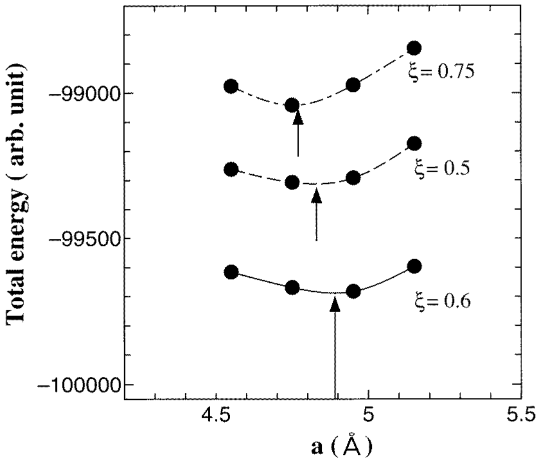

Figure 1. The total energy calculated for UC as a function of the lattice constant a for three values of the ratio of the C APW sphere radius to the U APW sphere radius, $\xi = R(\mathrm{C})/R(\mathrm{U})$. The arrows show the location of the minimum for each value of ξ.

As for the radii of the APW spheres involved in the muffin-tin approximation, there is no definite prescription as regards how to choose them. As usual, we select the spheres to be as large as possible without overlapping with each other. The relative sizes of the spheres, however, remain undetermined. To fix these free parameters, we employ the energy variational principle, i.e., we minimize the total energy by varying the radii of the U and C APW spheres for a given lattice constant. Then the total energy so determined is minimized as a function of the lattice constant. In figure 1 the total energy is represented as a function of the lattice constant a for three values of the ratio of the C APW sphere radius to the U APW sphere radius, $\xi = R(\mathrm{C})/R(\mathrm{U})$. As is shown in figure 1, it is clear that the total energy becomes minimum at $\xi = 0.6$ and $a = 4.89$ Å. As a result of this optimization, the U and C APW sphere radii have been determined as 3.056 25 a_0 and 1.833 75 a_0, respectively, at the equilibrium lattice constant $a_0 = 4.89$ Å. Details of the variational scheme for the APW spheres will be published elsewhere [8].

The value of a_0 is smaller by 1.4% than the experimental lattice constant 4.958 Å [1] that was used in the previous calculation [2]. The value of the ratio $\xi = 0.6$ is the same as was used previously.

The iteration process for solving the relativistic one-electron equation starts with a crystal charge density constructed by superposing the relativistic atomic charge densities for the neutral atoms U (Rn, $5f^36d^17s^2$) and C (He, $2s^22p^2$), where Rn and He stand for the electronic configurations of radon and helium, respectively. In the calculation for the atoms, the same exchange and correlation potential is used as for the crystal. The Rn core state except the $6p^6$ state for U and the He core state for C are assumed to be unchanged during the iteration, i.e., the frozen-core approximation is adopted for these states in the calculation for the crystal.

The crystal charge densities are constructed by taking 19 sampling $\boldsymbol{k}$-points which are distributed uniformly in the irreducible 1/48 part of the Brillouin zone. At each $\boldsymbol{k}$, about 200 relativistic APW basis functions are employed under the condition

$$|\boldsymbol{k} + \boldsymbol{K}_n| \leqslant 4.7(2\pi/a_0)$$

where the $\boldsymbol{K}_n$ are reciprocal-lattice vectors, and the angular momenta up to $l = 8$ are taken into account. Under these conditions, the total energy is found to converge well within a few mRyd by changing various parameters such as the number of sampling $\boldsymbol{k}$-points, the number of basis functions and the maximum angular momentum.

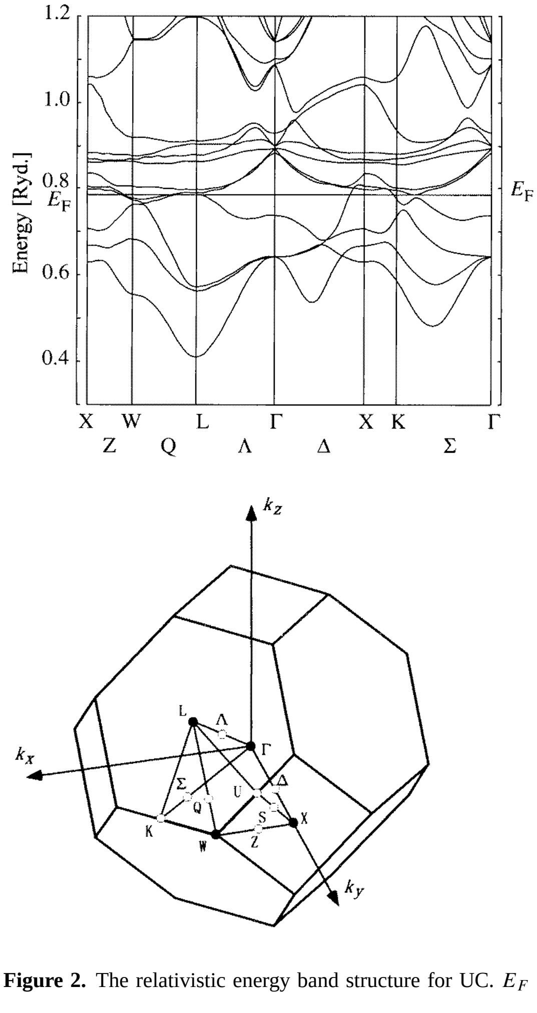

Figure 2. The relativistic energy band structure for UC. E_F shows the Fermi level.

3. Results and comparison with experiment

In figure 2, the energy band structure calculated for UC is shown. This result is qualitatively similar to the previous result [2]. Narrow bands which lie just above the Fermi level and are split into two subbands by the spin–orbit interaction are identified as the U 5f bands. The C 2p bands lie below the Fermi level and are hybridized significantly with the U 5f bands. The Fermi level is located at the bottom of the 5f bands and at the top of the 2p bands.

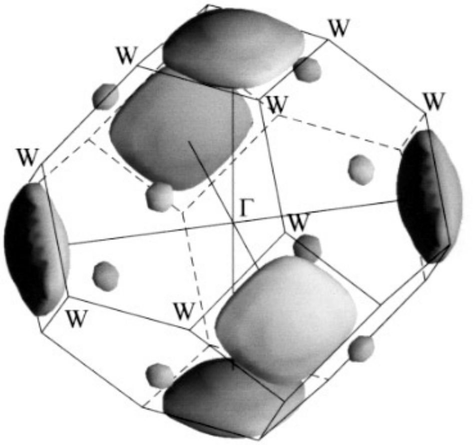

Figure 3. The hole sheets of the Fermi surface centred at the X and L points in UC.

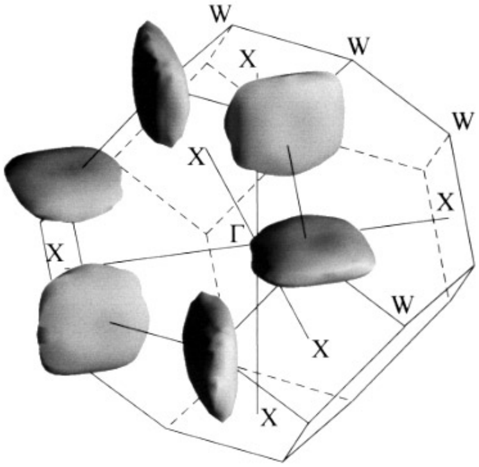

Figure 4. The electron sheets of the Fermi surface centred at the W points in UC.

Figure 2 shows that the Fermi surface consists of the hole sheets centred at the X points, the hole sheets centred at the L points and the electron sheets centred at the W points. These sheets of the Fermi surface are all small in size and closed. The hole sheets centred at the X and L points are shown in figure 3, and the electron sheets centred at the W points are shown in figure 4. The hole sheet centred at the L point is extremely small as compared with the hole sheet centred at the X point. The carrier numbers of holes and electrons are equal, i.e., UC is a semimetal. The hole sheet centred at the X point is larger in size than the electron sheet centred at the W point, because the number of electron sheets is double the number of hole sheets. The Bloch states on the electron sheets and on the hole sheets centred at the L points consist dominantly of U 5f states, while those on the hole sheets centred at the X points consist dominantly of C 2p states.

These results for the Fermi surface are similar to those of the previous calculation [2], though showing the existence of the small hole sheets centred at the L points required some delicacy in the previous calculation. This calculation predicts it clearly.

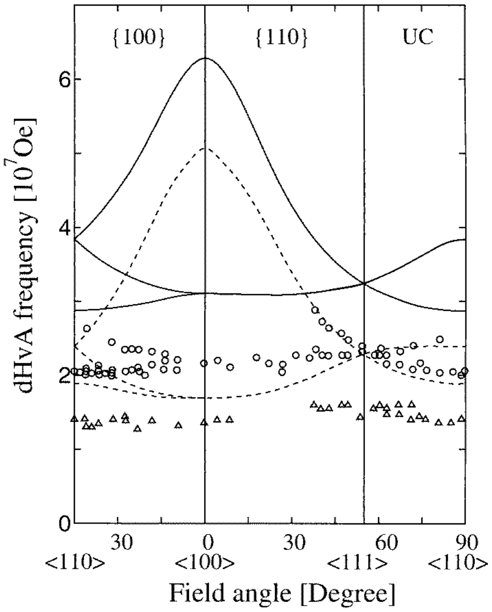

Figure 5. The angular dependence of the frequencies of the de Haas–van Alphen effect of UC. Circles and triangles show the experimental results obtained by Onuki *et al* [3]. Solid and broken lines show theoretical frequency branches which are calculated from the hole sheets centred at X and the electron sheets centred at W, respectively.

In order to compare with the experimental result for the dHvA effect, we calculate the extremal cross-sections of each sheet of the Fermi surface in various directions. The area of the extremal cross-section of the Fermi surface, A, is related to the dHvA frequency F by the formula $F = c\hbar A/2\pi e$. The theoretical result is shown together with the experimental result [3] in figure 5. In figure 5, the frequency branches which originate from the small hole sheets are centred at the L points, because they are extremely small in magnitude, i.e., of order 10^6 Oe. It is natural to assign the experimental branches with low frequencies to the electron sheets and to assign the experimental branches with high frequencies to the large hole sheets. Quantitative comparison reveals, however, that the theoretical frequencies are considerably larger in magnitude than the experimental frequencies. The frequency branches which correspond to the theoretical branches originating from the small hole sheets centred at the L point have not been observed.

4. Discussion

In this band calculation for UC, the U and C APW sphere radii and the lattice constant are determined so as to minimize the total energy within the framework of the local-density approximation. The degree of theoretical self-consistency has obviously been increased. In spite of this refinement of the calculation, the agreement between the theoretical dHvA frequencies and the experimental ones becomes poor. This result leads us to conclude that the electronic structure in the vicinity of the Fermi level may depend sensitively on the treatment of the exchange and correlation interaction between electrons. In other words, a defect of the local-density approximation manifests itself. This result may be related closely to the well-known theoretical result that the local-density approximation leads to an energy band gap of a semiconductor which is too small. To improve the band theory of the 5f-electron compounds on this point, a study on the basis of an optimized-effective-potential theory [9] is now in progress.

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