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Electronic band structure and properties of α -U (*)

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Résumé. — Utilisant une version relativiste de la méthode LAPW, nous présentons des résultats d'une étude détaillée de la structure de bande électronique ainsi que de quelques propriétés physiques de l'uranium dans la phase orthorhombique. Les densités d'états totales et partielles sont utilisées pour discuter les résultats de photo-émission de Veal et Lam. Les surfaces de Fermi des bandes prédominantes 6 et 7 présentent une symétrie élevée ainsi que quelques particularités topologiques (*nesting*) qui peuvent être comparées aux mesures actuelles d'effet dHvA d'Arko et Schirber. Le calcul du facteur de forme neutronique en présence d'un champ magnétique est en bon accord avec les mesures de Maglic *et al.*

Abstract. — We present some results of a detailed study of the electronic band structure and some physical properties of orthorhombic uranium using a relativistic version of the LAPW method. Total and projected (by orbital angular momentum) densities of states are used to discuss the photo-emission data of Veal and Lam. The Fermi surfaces of the dominant 6th and 7th bands are found to have high symmetry (and some nesting features) of interest to dHvA measurements being pursued by Arko and Schirber. The theoretical magnetic field induced neutron form factor is found to be in good agreement with the measurements of Maglic *et al.*

The anomalous physical properties of α -U have made it a subject of great interest for experimental and theoretical study. Unfortunately, its complex structure (orthorhombic with 4 atoms/unit cell) and the resulting lack of symmetry have made its theoretical study by conventional energy band methods exceptionally difficult and costly. Thus, while we were able to study the light and heavy actinide metals [1] in their cubic (high temperature) structures, the study of α -U awaited development of a computational scheme which was not only rapid and efficient but which avoided the so-called *asymptote problem* which plagued our earlier efforts.

In this paper we report some results of our relativistic energy band studies on α -U using a relativistic version of the linearized augmented plane wave method [2-3] (LAPW). This scheme has been successfully applied to the study of some actinide compounds, notably UGe₃ [4], UIr₃ [2], and URh₃ [5]. We have determined the energy band structure, density of states (DOS), orbital angular momentum projected DOS (for use in analysing the XPS experiments of Veal and Lam [6], detailed Fermi surface cross-sections in close collaboration with the work of Arko and Schirber [7] reported elsewhere at this conference), wavefunctions and magnetic field induced spin densities, neutron magnetic form factors (in close collaboration with and to understand the measurements of Maglic *et*

al. [8]), and generalized susceptibilities, $\chi(\mathbf{q})$ (for investigating possible electronically driven phonon anomalies and charge density waves). Because of space limitations we are able only to give a brief indication of this extensive work here. A full report is being prepared for publication elsewhere.

The three lattice parameters and the atomic positional parameter, u , were obtained by extrapolating the temperature dependent X-ray measurements of Barrett *et al.* [9] to the temperatures assuming that the system was cooled under pressure in order to avoid the anomalies associated with transitions at $T < 43$ K. Using these parameters ($a = 5.360$ Å, $b = 11.086$ Å, $c = 9.328$ Å, and $\mu = 0.102$) our model warped muffin tin potential was constructed in the standard overlapping charge density model using the Kohn-Sham-Gaspar ($\alpha = 2/3$) exchange. In our calculation we use the $f^3 d^2 s^1$ configuration. Different estimates including a pseudo-self-consistent model (as was used earlier for UGe₃) to determine an improved choice of configuration, indicate that a configuration in which roughly 0.25 to 0.5 electron are transferred from the f to the d levels is more appropriate. The calculated band structure was fitted with a Fourier series and used in our determinations of the Fermi surface, DOS and $\chi(\mathbf{q})$.

Figure 1 shows the $l = 3$ angular momentum projected DOS. We see a good deal of structure in the total DOS (not shown) arising from the lower symmetry and the hybridized set of s , p , d , and f bands. There is a rapidly increasing total DOS just above E_F

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Fig. 1. — The $l = 3$ projected DOS.

which has relevance to specific heat and susceptibility determinations in alloys of uranium and to optical absorption measurements. The $l = 3$ DOS correlates well with the XPS data of Veal and Lam [6] whereas the total DOS does not. This result is consistent with the fact that the higher angular momentum (more localized) f orbitals have much larger transition matrix elements. The large amount of $l = 3$ DOS above E_F indicates that this is the cause of the buildup in the total DOS in this same energy region. One might be tempted to attribute the occupied f-character (i.e. the f-DOS below E_F) to tailing caused by hybridization with the s, p, and d states. Although hybridization is present in this region of energy, many of the wave functions show nearly pure f-character and so hybridization alone is not responsible for the observed occupied f states but involved the formation of a true f-band.

Our calculated Fermi surface cross-sections are presented and discussed elsewhere in these proceedings. In the remaining space we present the magnetic field induced spin densities decomposed by l -value. The Fourier transform of these spin densities make the dominant contribution to the neutron magnetic form factor. Figure 2 compares the free ion f state charge density (broken curves) with the $l = 3$ decomposed radial density at (or close to) the Fermi energy, 1.135 Ry. All densities are normalized to unity over the r space shown (solid curves) so that they must be multiplied by their occupation. (The f contribution is roughly 80 % with the majority of the remainder comprised of d-character.) We find that the two $l = 0$ densities agree very closely, the $l = 1$ densities are slightly shifted for large r , and the $l = 2$ densities deviate only near the sphere radius. The major difference is in the $l = 3$ densities with the band result showing the large radial expansion expected for itinerant electrons in the lower half of the band. It is this expansion which is responsible for the contracted values of the magnetic neutron form factor seen by Maglic *et al.* [8].

Fig. 2. — Comparison of the free ion f state charge density with the $l = 3$ decomposed radial density at (or closed to) E_F .

Our results indicate that the f-electrons in α -U are not only quite itinerant in nature (and hence treatable by a band model), but are dominantly present in the experimental data.

References

- [1] FREEMAN, A. J. and KOELLING, D. D., *The Actinides : Electronic Structure and Related Properties*, A. J. Freeman and J. B. Darby ed. (Academic, New York) 1974.
- [2] KOELLING, D. D., *Proc. of the Second Intern. Conf. on the Electronic Structure of the Actinides*, J. Mulak, W. Suski and R. Troc (Ossolineum, Wroclaw, Poland) ; KOELLING, D. D. and ARBMAN, G. O., *J. Phys. F* 5 (1975) 2041.
- [3] ANDERSEN, O. K., *Phys. Rev. B* 12 (1975) 3060.
- [4] ARKO, A. J. and KOELLING, D. D., *Phys. Rev. B* 17 (1978) 3104.
- [5] ARKO, A. J. and KOELLING, D. D. (private communication).
- [6] VEAL, B. W. and LAM, D. J., *Phys. Rev. B* 10 (1974) 4902.
- [7] SCHIRBER, J. E., ARKO, A. J. and FISCHER, E. S., *Solid State Commun.* 17 (1975) 553.
- [8] MAGLIC, R. C., LANDER, G. H., MUELLER, M. H. and KLEB, R., *Phys. Rev. B* 17 (1978) 308.
- [9] BARRETT, C. A., MUELLER, M. H. and HITTERMAN, R. L., *Phys. Rev.* 129 (1963) 625.