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The calculated Fermi surface of osmium

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Abstract. On the basis of fully relativistic self-consistent band structure calculations a model of the Fermi surface of Os is suggested. The angular dependence of the extremal cross-section areas has been determined and found to be in good agreement with experimental data. Two additional groups of DHVA oscillations in the region of 200 MG associated with the large orbits on a hole 'monster' are predicted.

1. Introduction

Over the last three decades, the Fermi surfaces (FS) of most transition metals have been determined by using both theoretical and experimental methods (for a review, see Mackintosh and Andersen 1980). In many cases the results of band structure calculations and related Fermi surface parameters agree well with the data obtained by various experimental techniques.

Osmium, a hexagonal close-packed (HCP) 5d transition metal, presents some kind of exception to this rule. The Fermi surface of Os has been studied by magnetoresistance measurements (Alekseevskii *et al* 1968), the de Haas–van Alphen (DHVA) effect (Kamm and Anderson 1970), and the high-field induced-torque technique (Plachy and Dixon 1972). The data of all three experiments are consistent with each other, supporting the model of the Fermi surface suggested by Kamm and Anderson (1970). On the other hand, several non-self-consistent band structure calculations have been performed for Os using RAPW (Iyakutti *et al* 1977, Ramchandani 1980) and LMTO (Jepsen *et al* 1975) methods. Although different forms of exchange-correlation potentials have been employed, only qualitative agreement between theory and experiment has been achieved. Some discrepancies concerning Fermi surface topology have also appeared. Extremal cross-sections of the Fermi surface have been calculated only for high-symmetry directions.

The purpose of the present work was to study the Fermi surface of osmium in some detail in order to obtain more reliable information about its topology and dimensions. To achieve this, we performed calculations of self-consistent fully relativistic band structure and Fermi surface parameters of Os, including angular dependence of extremal cross-sections.

2. Method of calculation

We have employed, for band structure calculations, the fully-relativistic version of the linear muffin-tin orbitals (LMTO) method in atomic sphere approximation (ASA)

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developed by Andersen in 1975. Since the details of the method are described elsewhere (see e.g. Skriver 1980), only a brief account on its present version is given here. It implies using a two-component representation of the basis wavefunctions and a solution of the Dirac system of radial equations (see e.g. Nemoshkalenko *et al* 1983), in contrast to the usual relativistic treatment of solving the Pauli equation for one-component wavefunctions regarding the spin–orbital interaction as a perturbation. It seems to be extremely important in the case of osmium which has the maximum value of spin–orbit splitting of all 5d transition metals. To improve the accuracy of eigenvalue calculations, we constructed Hamiltonian and overlap matrices on MT orbitals up to $\kappa(l) = 3$ and included combined corrections in the ASA.

Self-consistent band structure calculations were performed on 936 k-points in the irreducible wedge (1/24 part) of the Brillouin zone for an HCP lattice. The exchange–correlation potential in the form suggested by von Barth and Hedin (1972) was used. Lattice parameters $a = 2.735$ Å and $c = 4.324$ Å giving a c/a ratio of 1.58 were taken in accordance with the low-temperature experimental values quoted by Pearson (1958).

To calculate extremal areas of the Fermi surface, we employed a relatively simple linear interpolation procedure of cross-section determination based on the well-known tetrahedron method, assuming linear dependence of energy between k-points. Cyclotron masses m have been calculated using the well-known formula (in atomic units)

$$m/m_0 = (1/\pi) \oint_{FS} \mathrm{d}l/v_\perp$$

where $v_\perp$ is the component of velocity vector $\boldsymbol{v}$ in the cross-section plane, and integration is over the particular Fermi surface sheet.

An empty lattice test on a BCC lattice using approximately 500 tetrahedra gave us a maximal relative error of less than 1% for extremal cross-sections and less than 5% for cyclotron masses.

3. Results and discussion

3.1. Energy band structure

Our band structure of osmium, calculated as outlined in the previous section, is shown in figure 1. It appears to be much more similar to that obtained by Jepsen *et al* (1975) using a non-self-consistent LMTO method with standard Slater X_α exchange–correlation potential rather than to those obtained by the RAPW method with different forms of potential. In fact, the only major difference between our band structure and that calculated by Jepsen *et al* (1975) is at the Γ and L points, where the tenth and seventh bands respectively cross the Fermi level. In our band picture these eigenvalues lie much lower on the energy scale, as can be easily seen when comparing the two band structures.

Dealing with RAPW calculations it should be noted that in the work by Iyakutti *et al* (1977) two alternative forms of the exchange–correlation part of the potential were employed: using the Wigner formula (V_1) and using the Overhauser (1971) expression (V_2). It is stated by the authors that in the first case results quite similar to that of Jepsen *et al* (1975) were obtained. Calculation results with potential V_2 are completely different. The width of the conduction band has enormously increased, affecting the whole Fermi surface toplogy. As to the calculation by Ramchandani (1980), the RAPW method with Slater X_α exchange–correlation potential was employed. Results very similar to the

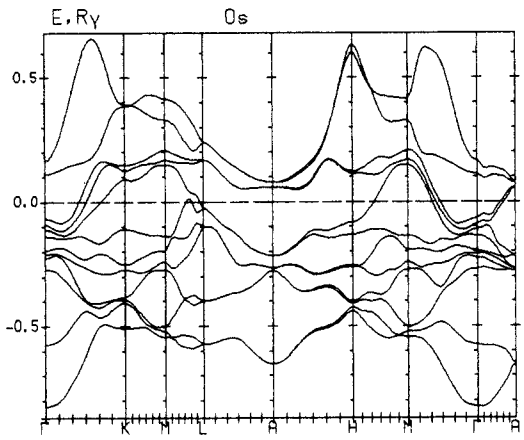

Figure 1. Fully relativistic self-consistent energy band structure of Os.

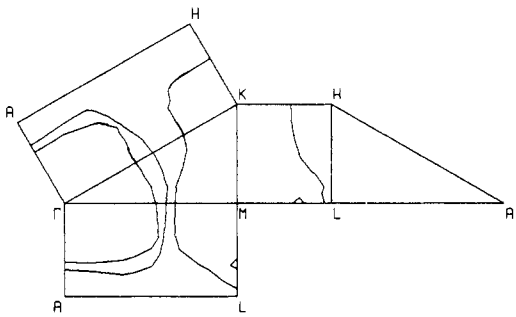

Figure 2. Cross-sections of Os Fermi surface in the irreducible part of the Brillouin zone.

calculation of Iyakutti *et al* (1977) with V_2 potential were obtained. In this case the calculated conduction bandwidth is also too high. For example, the tenth band does not even intersect the Fermi level in the ΓK direction, obviously causing incorrect Fermi surface topology.

To proceed further, let us now consider a Fermi surface model suggested on the basis of our band structure calculations and compare it with other theoretical results and experimental data.

3.2. Fermi surface model

Four sheets of osmium Fermi surface are clearly seen in figure 1. Cross-sections of FS by Brillouin zone faces are shown in figure 2, while 3D computer-generated drawings of these sheets originating from different bands are presented in figures 3–5. They are: two nested electron-type ellipsoids e_9 and e_{10} centred at Γ, a network of multiply connected toroidal hole-type pieces h_8 (the so-called 'monster') reflecting the hexagonal symmetry of the lattice and a set of small hole-type ellipsoids h_7 centred on the line LM of the face of the Brillouin zone.

As to the results of other calculations, along with these sheets Jepsen *et al* (1975) and Iyakutti *et al* (1977) have found the occurrence of a hole ellipsoid at the centre of the Brillouin zone due to the crossing of the Fermi level by the tenth band near Γ. Moreover, an additional hole sheet at L has been found by Jepsen *et al* (1975).

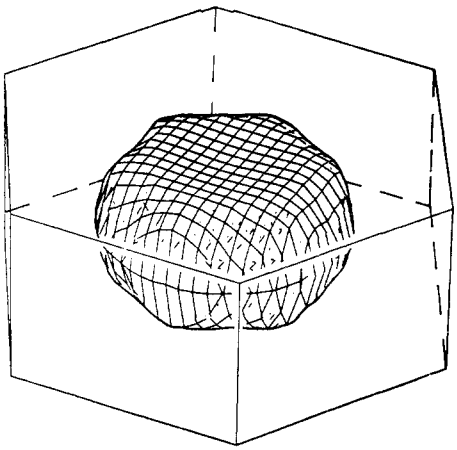

Figure 3. Electron ellipsoid e_9 in the first Brillouin zone.

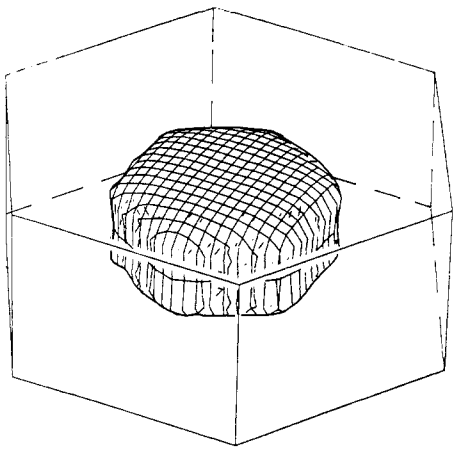

Figure 4. Electron ellipsoid e_{10} in the first Brillouin zone.

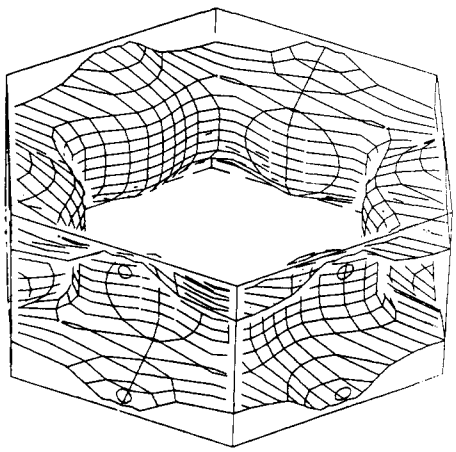

Figure 5. Hole surface h_8 in the first Brillouin zone. Hole ellipsoids h_7 cross-sections are also shown.

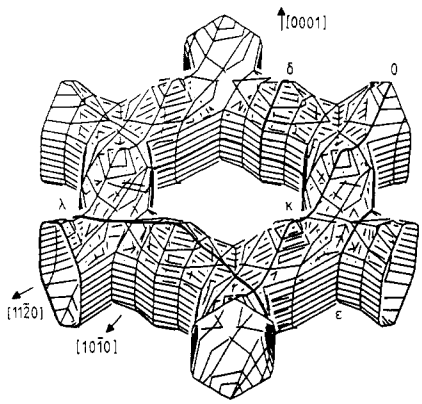

Figure 6. Hole surface h_8 in the extended zone scheme. Greek letters denote various extremal orbits. 0 denotes open orbit in the $\langle 10\bar{1}0\rangle$ direction.

The osmium Fermi surface was first studied experimentally by investigation of its galvanomagnetic properties (Alekseevski *et al* 1968). On the basis of the magnetoresistance data obtained it was concluded by the authors that the Fermi surface is open along the [10$\bar{1}$0] and two equivalent directions lying in the basal plane. In addition, an open-orbit path along the [0001] direction appeared in strong ($H > 40$ kG) magnetic fields. These results have since been confirmed using an induced-torque method (Plachy and Dixon 1972).

A more detailed study of the osmium Fermi surface was performed in a DHVA experiment by Kamm and Anderson (1970). The DHVA frequencies of two groups were

Table 1. Extremal cross-section areas and cyclotron masses in high symmetry directions.

FS sheet	Direction and osc. type	Extremal areas (MG)					Cyclotron masses (m/m_0)		
		Theory	Expt. (DHVA)	Theory (Jepsen)	Theory (Iyakutti V_1, V_2)		Theory	Expt. (DHVA)	Theory (Jepsen)
h_7	[0001]α	0.73	2.99	5.70			0.09	0.23	0.20
	[10$\bar{1}$0]α	0.40	1.43	2.40			0.05	0.13	0.10
	[11$\bar{2}$0]α		2.24	3.20				0.18	0.10
	[11$\bar{2}$0]α	0.34	1.54	2.10			0.07	0.15	0.10
h_8	[0001]κ	291.8					1.38		
	[10$\bar{1}$0]δ	106.3	110.0	111.0			0.76	1.49	1.00
	[10$\bar{1}$0]λ	242.0					1.55		
	[11$\bar{2}$0]δ	104.3	109.0	109.0			0.67	1.10	0.87
	[11$\bar{2}$0]ε	66.5	68.0	65.0			0.78	1.10	0.99
e_9	[0001]γ	206.3	205.3	207.0	169.8	209.6	0.80		1.09
	[10$\bar{1}$0]γ	160.0	160.0	163.0	227.5	233.4	0.95		1.30
	[11$\bar{2}$0]γ	169.3	168.0	175.0	177.3	153.8	1.08		1.49
e_{10}	[0001]β	153.2	153.0	152.0			0.84	1.25	0.97
	[0001]β	148.4	148.0	149.0	132.9	135.5	0.70	1.25	0.90
	[10$\bar{1}$0]β	122.7	124.8	124.0	190.3	151.7	0.84	1.26	1.12
	[11$\bar{2}$0]β	132.8	133.8	133.0	165.0	136.3	0.93	1.26	1.23

detected. Since at that time there were no band structure calculations for osmium they used a band structure calculated by Mattheiss for rhenium with a Fermi level shift appropriate to osmium to explain their results. According to their interpretation, lower-frequency oscillations labelled α result from a set of small ellipsoids centred at the face of the Brillouin zone. The highest frequencies observed, β and γ, represent two ellipsoids centred at Γ. Since the forms of these oscillations are very similar it means that one ellipsoid is nested within the other. Intermediate-frequency oscillations δ and ε lying mainly in the basal plane result from a complicated multiply-connected surface.

Taking into account all these arguments it could be concluded that in contrast to the previous calculations our calculated Fermi surface topology is the same as predicted by various experiments.

The theoretical and experimental extremal areas of the Fermi surface and cyclotron (effective) masses are given in table 1 for the principal symmetry directions. The greek letters correspond to the original notation used by Kamm and Anderson (1970).

3.3. Closed orbits and oscillation allocation

Simplest to discuss are the α group of oscillations attributed to small ellipsoids. It could be easily seen from figure 5 that these ellipsoids are centred between points L and M on the centre line of the HCP Brillouin zone. Because of the small dimensions the accuracy of our extremal area determination using a linear interpolation method is not high.

In order to determine complete osmium Fermi surface dimensions we have calculated the angular dependence of extremal cross-sections of large pieces, which is presented in figure 7 along with higher-frequency DHVA oscillations.

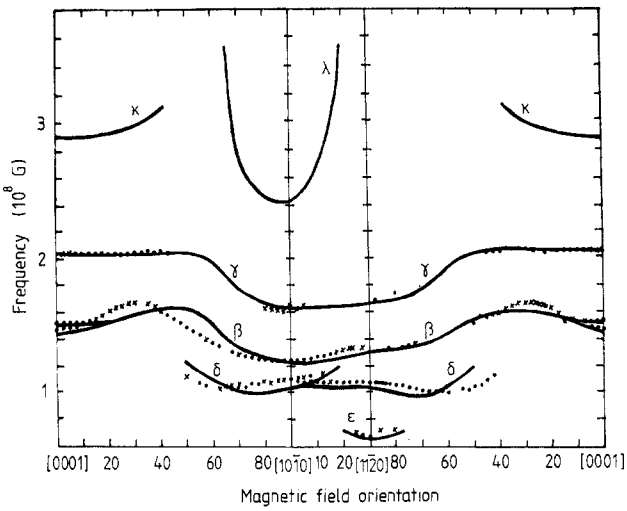

Figure 7. Angular dependence of calculated (full lines) and measured (circles and crosses) extremal cross-sections. Experimental points denoted by circles are the principal components of DHVA oscillations, while those denoted by crosses have been determined from beating oscillations.

Allocation of γ and β frequencies to the extremal orbits on the surfaces of e_9 and e_{10} ellipsoids centred at Γ is straightforward. Agreement between calculated angular dependence of extremal cross-sections and angular variations of the respective DHVA frequencies is rather good. Two extremal cross-sections of smaller surface along the [0001] direction clearly indicate that this ellipsoid is waisted, as shown in figure 4.

As to the intermediate-frequency oscillations δ and ε, the agreement is also good. These oscillations result from multiply-connected hole 'arms' h_8 lying in the basal plane, with ε results from the minimum in [11$\bar{2}$0] and equivalent directions and δ from the maximum section of this sheet just as predicted by Kamm and Anderson and as can be clearly seen from figure 6. However, there are additional giant orbits on this surface not observed experimentally. An extremal orbit on the inner diameter of the hole 'monster' would inevitably cause oscillations labelled κ in the [0001] direction, while an extremal orbit connecting two opposite 'arms' would result in λ oscillations situated mainly in the [10$\bar{1}$0] direction. Further DHVA experiments in the range of 200–300 MG would be desirable to detect these orbits.

3.4. Open orbits and the possibility of magnetic breakdown

As was already mentioned above, the magnetoresistance measurements of Alekseevskii *et al* (1968) have detected open-orbit trajectories along the [10$\bar{1}$0] direction. This was confirmed by the induced-torque measurements of Plachy and Dixon (1972). According to our FS model, such open orbits definitely exist on the surface of the hole 'monster' as shown in figure 6.

In addition, magnetoresistance data revealed the appearance of open orbits along the hexagonal axis in strong magnetic fields. This is impossible according to our model and data from the DHVA experiment since such open orbits would require the sheet h_8

to be in contact with the AHL plane of the Brillouin zone. However, the [0001] direction would appear to be open if magnetic breakdown through this plane occurs which could take place in strong magnetic fields. This situation is quite common among HCP transition metals and has been discussed e.g. by Jepsen (1975) for Ti.

4. Conclusions

On the basis of fully relativistic band structure calculations a model of the osmium Fermi surface is suggested. Calculated Fermi surface topology and dimensions are in good agreement with experimental DHVA, magnetoresistance and induced-torque data. Closed orbit allocation to the measured extremal cross-sections has been made along with determination of their angular dependence. In addition, two groups of DHVA oscillations associated with orbits on the hole 'monster' have been predicted. Open orbits in the [10$\bar{1}$0] and equivalent directions explicitly follow from calculated FS topology, while those in [0001] imply that magnetic breakdown occurs as in some other HCP metals.

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