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Journal of Physics and Chemistry of Solids 65 (2004) 2011–2016
JOURNAL OF PHYSICS AND CHEMISTRY OF SOLIDS
www.elsevier.com/locate/jpcs

Refining Fermi surface topologies from ab initio calculations through momentum density spectroscopies

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Abstract

It is well known that the agreement between the Fermi surface topologies predicted by ab initio electronic structure calculations and experiment can often be brought into much better agreement through small rigid-band-like shifts. A new method for refining these calculations using experimental data containing Fermi surface information, based on a rigid-band-like fitting approach is presented. In this method, experimental data from different methods can be combined to refine and deliver a 'tuned' bandstructure, allowing an investigation of FS nesting properties, a quantitative comparison between experiment and calculation, and highlighting the origin of inconsistencies. Results of the application of this method to positron annihilation experiments in vanadium are presented, showing significant improvement over the ab initio calculation. In order to demonstrate the versatility of this fitting method, it has been applied to a combination of positron annihilation measurements and magnetic Compton scattering experiments in ferromagnetic nickel.

PACS: 71.18.+y; 78.70.Bj; 78.70.Ck; 71.20.Be

Keywords: C. Ab initio calculations; C. Position annihilation; D. Fermi surface; D. Electronic structure

1. Introduction

In many instances an accurate description of the Fermi surface (FS) topology is necessary for an understanding of the physical properties of a metallic system. For example, the propensity of a system towards various forms of magnetism, superconductivity or other kinds of order (including charge- or spin-density waves and compositional short-range order in substitutionally disordered random alloys), is often strongly influenced by the shape of the FS (see for example [1–3]). While density-functional electronic structure calculations have proved enormously powerful in providing descriptions of this electronic structure, it is well known that even in simple systems small inaccuracies can lead to incorrect predictions for these FS topologies (for examples of such systems relevant to the current work, see [4–10] and references therein). Such inconsistencies with experimental measurements have led to the application of various (mostly empirical) procedures to bring the theory into line with experiment [11–14].

Our recent work has focused on delivering (often unique) information on the FS topologies of a variety of systems where that topology is thought to drive particular physical phenomena. In these systems which often possess complex multi-band FS topologies, our experimental measurements are usually complemented by ab initio electronic structure calculations to aid the interpretation. It is frequently the case that much better agreement between the experimentally derived and theoretically predicted FS topologies can be obtained by simply rigidly shifting the bands by small amounts with respect to the Fermi level. In this paper we present the results of a new method aimed at taking an existing calculation of the electronic band structure and 'tuning' it in a rigid-band manner to the experimentally obtained FS. Its novelty and virtue lies in its versatility in including relevant data from a variety of experimental probes sensitive to the FS. These experimental probes include quantum oscillatory methods [15] such as the de Haas-van Alphen effect (dHvA, which can, in favourable

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doi:10.1016/j.jpcs.2004.08.013

circumstances, deliver very precise information about extremal FS areas), and spectroscopies such as positron annihilation [16], Compton scattering [17] and angle-resolved photoemission (ARPES, see for example [18]).

We will show that such a simplistic rigid-band approach applied to data obtained from positron annihilation and magnetic Compton scattering experiments can 'tune' the predicted FS topologies, and demonstrate that their extremal areas become consistent with those independently measured in quantum oscillatory experiments. Such refined bandstructures are essential for investigating the nesting properties of the FS, usually identified through singularities in the wavevector-dependent susceptibility [19,20]. In addition, with this procedure we also obtain a description of the full three-dimensional FS, complementary to that obtained by tomographic reconstruction from positron annihilation experiments [1].

To the best of our knowledge, the only other attempt to directly obtain the FS from positron annihilation data through a fitting procedure involved the parameterization of the FS in terms of a model topology specific to group VI metals [21].

2. Experimental methods

Two-dimensional angular correlation of electron–positron annihilation radiation (2D-ACAR) can be used for experimentally investigating the occupied momentum states, and hence FS topologies [16]. A 2D-ACAR measurement yields a 2D projection (integration over one dimension) of an underlying two-photon momentum density (TPMD), $\rho^{2\gamma}(\mathbf{p})$

$$\rho^{2\gamma}(\mathbf{p}) = \sum_{\mathrm{OCC},j,\mathbf{k}} \left| \int \mathrm{d}\mathbf{r} \sqrt{\gamma(\mathbf{r})} \psi_{\mathbf{k},j}(\mathbf{r}) \psi_{+}(\mathbf{r}) \exp(-\mathbf{i}\mathbf{p}\cdot\mathbf{r}) \right|^2 = \sum_{j,\mathbf{k},\mathbf{G}} n^j(\mathbf{k}) |C_{\mathbf{G},j}(\mathbf{k})|^2 \delta(\mathbf{p}-\mathbf{k}-\mathbf{G}), \quad (1)$$

where $\psi_{\mathbf{k},j}(\mathbf{r})$, and $\psi_{+}(\mathbf{r})$ are the electron and positron wave functions, respectively, and $n^j(\mathbf{k})$ is the electron occupation density in $\mathbf{k}$-space in the jth band, and $\gamma(\mathbf{r})$ is the so-called enhancement factor which takes account of electron–positron correlations (and would be unity in the independent particle model) [22]. The $C_{\mathbf{G},j}(\mathbf{k})$ are the Fourier coefficients of the interacting electron–positron wave function product and the delta function expresses the conservation of crystal momentum. $\rho^{2\gamma}(\mathbf{p})$ contains information about the occupied electron states and their momentum, $\mathbf{p}=\hbar(\mathbf{k}+\mathbf{G})$, and the FS is reflected in the discontinuity in this occupancy at the Fermi momentum, $\mathbf{p}_\mathrm{F}=\hbar(\mathbf{k}_\mathrm{F}+\mathbf{G})$.

A related technique is Compton scattering [17]. Here the electron momentum density (EMD), $\rho(\mathbf{p})$, is probed and a double integral of it is obtained (Compton profile, CP):

$$J(p_z) = \int\int \rho(\mathbf{p}) \mathrm{d}p_x \mathrm{d}p_y. \quad (2)$$

In a spin-polarized system, the total EMD is composed of (different) contributions from spin-up and spin-down electrons, $\rho(\mathbf{p})=\rho_\uparrow(\mathbf{p})+\rho_\downarrow(\mathbf{p})$, where $\rho_\uparrow(\rho_\downarrow)$ represents the spin-up (spin-down) EMD. Magnetic Compton Scattering [23] provides a double integral of the spin density in momentum space (magnetic Compton profile, MCP):

$$J_\mathrm{mag}(p_z) = \int\int (\rho_\uparrow(\mathbf{p}) - \rho_\downarrow(\mathbf{p})) \mathrm{d}p_x \mathrm{d}p_y. \quad (3)$$

We will demonstrate how TPMDs can be used on their own or in conjunction with magnetic EMDs to 'tune' the bandstructure to the experimental FS.

3. Rigid-band-like fitting procedure

The simplest imaginable procedure for bringing the FS topology predicted by an electronic structure calculation into better agreement with experiment is to shift the Fermi level. This is routinely applied by band theorists when comparing their results with quantum oscillatory measurements (see for example [11]). While such a procedure will not guarantee the correct occupied volume of the Brillouin zone (that is to say, the correct number of electrons), if the FS shape is basically correct then it is frequently a useful first step in understanding the successes and shortcomings of the calculation. Our method of 'tuning' ab initio bandstructure calculations to experimental data, in order to obtain a description of the system closer to reality, is based on such a rigid-band approach.

We used the electronic bandstructure and wavefunctions originating from self-consistent calculations using the linearized muffin-tin orbital (LMTO) method, within the atomic sphere approximation (ASA), including combined-correction terms [24]. The exchange-correlation part of the potential was described within the local density approximation (LDA) to the exchange-correlation functional (Hedin-Lundqvist [25] for the non-magnetic case, and Gunnarson-Lundqvist [26] for the spin-polarized calculations). For the TPMD, electron–positron correlation was included using the prescription of Jarlborg and Singh [22]. A full description of the method including the subsequent calculation of the EMD and TPMD is given in [27]. We assign a cut-off energy (which acts as a Fermi energy) to each band crossing the Fermi level, and include contributions to the momentum density up to that cut-off. In this way, one can think of the bands being shifted rigidly up and down with respect to the Fermi level. Since only shifts in the bands crossing the Fermi level result in a change in the momentum densities (and indeed any quantity associated with the topology of the FS) we only allow the contribution of these bands to vary. For each shift of each relevant band the corresponding quantity (examples of which are listed below) is calculated and then compared to the experimentally obtained one. These experimental quantities can take a variety of forms and could include