Properties of the Nearly Free Electron Superconductor Ag$_5$Pb$_2$O$_6$
Inferred from Fermi Surface Measurements

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Abstract

We measured the Fermi surface of the recently discovered superconductor Ag$_5$Pb$_2$O$_6$ via a de Haas-van Alphen rotation study. Two frequency branches were observed and identified with the neck and belly orbits of a very simple, nearly free electron Fermi surface. We use the observed Fermi surface geometry to quantitatively deduce superconducting properties such as the in-plane and out-of-plane penetration depths, the coherence length in the clean limit, and the critical field; as well as normal state properties such as the specific heat and the resistivity anisotropy. ©2006 Elsevier Science. All rights reserved

Key words: Ag$_5$Pb$_2$O$_6$, Fermi surface, penetration depth, anisotropy

PACS: 74.70.Dd, 74.25.Jb, 71.18.+y

1. Introduction

Superconductivity has recently been discovered in Ag$_5$Pb$_2$O$_6$ [1]. The material is the first layered silver oxide superconductor, and one of only a handful of known non-elemental type-I superconductors. While the normal state of Ag$_5$Pb$_2$O$_6$ is a good metal, it exhibits a quasi-$T^2$ dependence of the electrical resistivity up to room temperature which arises from a yet unidentified scattering mechanism [2]. There have been several band structure calculation on this material with contradictory results [3,4,5]. To elucidate these issues, we experimentally determined the Fermi surface (FS) using the de Haas-van Alphen (dHvA) effect and calculated the resulting semiclassical values for several superconducting and normal state parameters.

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Fig. 1. The experimentally derived FS of Ag$_5$Pb$_2$O$_6$ inside the first Brillouin zone.

2. Experimental Fermi Surface

Our dHvA experiment, reported in full in reference [6], revealed that the FS of Ag$_5$Pb$_2$O$_6$ is nearly free electron-like, as shown in Fig. 1. Similar to the noble metals, the FS occupies half the Brillouin zone and punches through its boundary to form neck orbits. The effective masses for the neck and belly orbits were found to be $m^*/m_e = 1.25 \pm 0.1$ and $1.1 \pm 0.2$. 

Preprint submitted to Elsevier Science
The FS is in very good agreement with band structure calculations by Oguchi [5] and those by Shein and Ivanovskii [4], but differs from those by Brennan and Burdett [3]. However, Oguchi [5] predicts a much lighter neck orbit \((m^*/m_e = 0.68)\) than we see in experiment, while his belly orbit mass matches rather well. The only way to reconcile that calculation with our data is to invoke very anisotropic electron-phonon coupling over the FS.

3. Parameterisation and Calculations

In order to calculate several normal state and superconducting parameters from our dHvA data, we need to parameterise the experimental FS and band structure. The lattice parameters are \(a = 5.93\ \text{Å} \) and \(c = 6.41\ \text{Å}\), and we obtain a very good fit to our data (with deviations between the observed and the fitted dHvA frequencies of 0.1 kT or less at all angles) by using

\[
E = \frac{\hbar^2}{2m^*}(k_x^2 + k_y^2) - 2t_\perp \cos ck_z
\]

with \(m^* = 1.2m_e, \ t_\perp = 0.157\ \text{eV}\), and a Fermi energy \(E_F = 0.656\ \text{eV}\). While the \(c\)-axis dispersion formally has a tight binding form, it should rather be thought of as “loose binding” since \(t_\perp\) is of order \(E_F\). This energy dispersion gives the same dHvA mass \(m^* = 1.2m_e\) for both neck and belly orbits, in line with experiment. The Fermi volume in this model gives an electron density of \(n = m^*E_F/\pi\hbar^2\) which agrees to within 1% with the stoichiometric expectation of one electron per unit cell.

Various parameters can now be calculated from the band structure as surface or volume integrals over the Fermi surface. Some, but not all, of these can be performed analytically. Table 1 lists the results in the limit \(T \to 0\).

First, the specific heat coefficient is proportional to the density of states at \(E_F\); specifically here we get \(\gamma = \pi k_B^2a^2m^*N_A/2\sqrt{3}\hbar^2\). The penetration depth can be calculated from the semiclassical Chandrasekhar-Einzel integral [7], the results are a very familiar in-plane value \(\lambda_{\parallel}^2 = n\varepsilon^2\mu_0/m^*\) and a \(c\)-axis penetration depth \(\lambda_{\perp}^2 = 2\varepsilon^2\mu_0m^*e^2/\pi\hbar^4\). The in-plane and \(c\)-axis coherence lengths are estimated from the RMS values of the Fermi velocities via \(\xi \simeq 0.18\ h_F/k_BTc\). They are much larger than the typical mean free path, implying that Ag\(_x\)Pb\(_{2}\)O\(_{6}\) is almost always in the dirty limit (see also [1]). We nevertheless list the clean limit critical field in Table 1 also. The normal state conductivity tensor can be calculated as a standard Fermi surface integral [8], where we assume a constant mean free path.

4. Discussion

The values for \(\xi\) and \(\lambda\) identify Ag\(_x\)Pb\(_{2}\)O\(_{6}\) as type-I (in the clean limit). The resistivity anisotropy \(\rho_\perp/\rho_\parallel\) comes out smaller than measured [2], indicating either a very anisotropic scattering rate or uncertainties about the geometry factor in those measurements. We believe that Ag\(_x\)Pb\(_{2}\)O\(_{6}\) is the first example of a nearly free electron superconductor, and it has the “most spherical” FS of all known superconductors. We now understand the normal state of Ag\(_x\)Pb\(_{2}\)O\(_{6}\) rather well, but the anomalously strong \(T^2\) term in the resistivity remains mysterious.

Acknowledgments We would like to thank J. Fletcher, G. G. Lonzarich, and P. Pyykkö for useful discussions. C.B. acknowledges the support of the Royal Society.

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Table 1

| From FS | Direct [1,2] |
|---------|--------------|
| \(\gamma\) | 3.6 mJ/K\(^2\) mol(f.u.) | 3.4 mJ/K\(^2\) mol(f.u.) |
| \(\lambda\) | 81 nm/116 nm | — |
| \(\xi\) | 8.4 \(\mu\)m/5.9 \(\mu\)m | — |
| \(B_c\) | 0.33 mT | 0.2 mT |
| \(\rho_\perp/\rho_\parallel\) | 2.1 | 6.5 |

"Fermi volume in this model gives an electron density of... specifically here..."
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