

High-field de Haas–van Alphen studies of the Fermi surfaces of LaMIn_5 ($M=\text{Co}, \text{Rh}, \text{Ir}$)

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We report measurements of the de Haas–van Alphen effect on a series of compounds, LaMIn_5 ($M=\text{Co}, \text{Rh}, \text{Ir}$). The results show that each of the Co and Ir Fermi surfaces (FSs) exhibits some portions that are two dimensional and some portions that are three dimensional. The most two-dimensional character is exhibited in LaCoIn_5 , less two-dimensional behavior is seen in LaIrIn_5 , and no part of Fermi surface of LaRhIn_5 is found to have a two-dimensional character. Thus the two dimensionality of portions of the FSs is largely determined by the d character of the energy bands while all of the effective masses remain ≤ 1.2 . This fact has implications for the causes of the heavy fermion nature of superconductivity and magnetism in the Ce-based compounds having similar composition and structure.

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I. INTRODUCTION

Prior to the discovery of magnetic superconductors, it was believed that the inclusion of magnetic rare-earth (RE) atoms in a material would effectively prevent the formation of a superconducting state. Weak ferromagnetic ordering was enough to stop the formation of Cooper pairs.

In the mid-seventies rare-earth ternary compounds were discovered that had both antiferromagnetic ordering and a superconducting state.^{1,2} In these materials ferromagnetic ordering via exchange interactions broke apart Cooper pairs; however, long-range antiferromagnetic order was essentially invisible to the formation of superconducting charge carriers.

Not long after the discovery of these antiferromagnetic superconductors, the first heavy fermion superconductors were discovered.³ These materials also exhibit weak antiferromagnetic ordering with the addition that the effective mass of the charge carriers are much larger than in normal metals, typically 10 to a 100 times the electron mass.

In order to fully explore the probable causes of these measured phenomena, it is necessary to find clean isostructural systems where the effects of chemical doping changes can be measured along with other thermodynamic changes. One such system is the rare earth-115 family consisting of dozens of compounds and intermediate dopings.

For the present work we have focused on one subset of this RE-115 family, the REMIIn_5 ($\text{RE}=\text{La}$ or Ce ; $M=\text{Co}, \text{Rh},$ or Ir). One of the most interesting members of this family is CeCoIn_5 since the transition from the superconducting state to normal state at high magnetic fields is first order which offers the possibility that this material is the first to exhibit a

Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting state.⁴

CeRhIn_5 has an antiferromagnetic ground state that can be driven into a superconducting state with the application of pressure.⁵ In CeIrIn_5 the resistive and magnetic transitions in zero field do not occur at the same temperature.

Gauging to what extent the $4f$ electrons, contributed by the Ce atoms, determine the various properties of these materials requires the slow removal of these electrons from the lattice. Measurements on $\text{Ce}_x\text{La}_{1-x}\text{In}_5$ were the subject of another study. It was found that even 10% dilutions of the magnetic $4f$ electrons would effectively remove the heavy fermion and superconducting properties of these materials. The conclusion is that magnetism is at the heart of understanding the rich physics of this system.

To date there has only been a single published study of the Fermi surface (FS) of the LaMIn_5 materials and that study only concerned LaRhIn_5 . It was found (and we confirmed this) that the CeRhIn_5 FS is almost identical to LaRhIn_5 . This is taken as an indication that the f electrons are localized in CeRhIn_5 and thus are not a major contributor to the FS topology.

CeCoIn_5 and CeIrIn_5 on the other hand are thought to exhibit some delocalization of the f electrons. This would suggest that the removal of the f electrons from the material would have a significant effect on the FS. We have measured the de Haas–van Alphen (dHvA) effect in LaCoIn_5 and LaIrIn_5 . We find that, while there are some differences, the general character of the FS is preserved under the complete removal of the f electron. This indicates that it is the d electrons in these materials that contribute most to the FS with

the f electrons contributing only small changes and a significant renormalization of the effective mass of the conduction electrons.

Because electrons on the FS are thought to play a role in all of these exotic properties, there has been considerable effort made in measuring their FSs over the past few years. The first measurements on the $CeMIn_5$ series were done on the Co-based material,⁶ then on the Rh material,⁷ and finally on the Ir material.⁸ In every case there is evidence that at least parts of the FSs are cylindrical with axes along the ΓZ direction of the Brillouin Zone, similar to high-temperature superconducting cuprates. Both $CeMIn_5$ and $LaMIn_5$ can be described as having a layered structure with the layers viewed as alternating La-In and M-In planes. If electrical conduction only takes place in two dimensions in a material, the electronic structure of the material is confined to these two dimensions and the FS will be perfectly cylindrical. Consequently, the dHvA measurements that determine the cross sectional areas of the FS perpendicular to an applied field would have an angular dependence of the measured frequencies proportional to $1/\cos(\theta)$, where θ is the angle between the direction of the applied field and the direction perpendicular to the layers. When there is conduction between the planes, other three-dimensional (3D) pieces of FS can exist, and the nearly cylindrical FS will undulate in area along the $\theta=0$ direction. Moreover, several frequencies may appear to obey the $1/\cos(\theta)$ rule over a limited angular range.

It is known that the heavy fermion properties of Ce-based compounds are due to delocalization of the cerium $4f$ electrons, forming bands with f character. However, the delocalization may not be complete, and energy-band calculations of the real ground state only recently have been performed. For this reason, we have undertaken the measurements of the FS of the La-based $LaMIn_5$ ($M=Co, Rh, Ir$) compounds that have the same structure as the Ce-based ones but have no $4f$ electrons. The results give information about the effects of the different d bands and a starting point for future energy-band calculations that should be able to describe these FSs correctly.

II. EXPERIMENTAL PROCEDURE AND DATA ANALYSIS

All of the results reported here are from data taken at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee, Florida. Most of the measurements were made using a metal film cantilever in a rotating sample holder. However, some of the results were checked using balanced pickup coils and magnetic-field modulation. (These techniques are described in many previous papers, including our own, e.g., see Ref. 6.) The frequency of the dHvA effect was measured for two different samples of each of the three values of M (Co, Rh, Ir) using different measurement techniques. Field modulation using balanced pickup coils is more sensitive to the high frequencies while the metal film torque measurement is best for detecting the lowest frequencies. The samples and the cantilever or pickup coils were immersed either in liquid 4He that could be pumped to 1.5 K or in liquid 3He with a base temperature of approximately 0.5

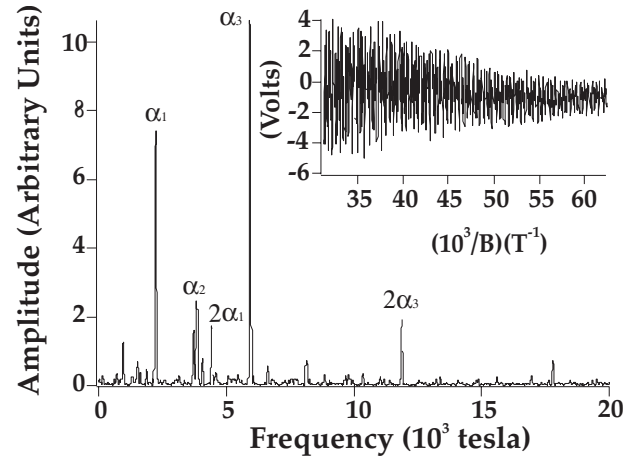


FIG. 1. Fourier transform of the dHvA data for $LaIrIn_5$. The raw data plotted as a function of reciprocal field is shown in the inset.

K. All of the data analyzed were performed on data taken in the field range of 15–33 T in one of the resistive magnets at the NHMFL.

In Fig. 1 we show data from $LaCoIn_5$ with the field perpendicular to the planes along the c axis. In the main graph a Fourier analysis of the data for fields between 15 and 33 T is shown while in the inset the actual raw data over this field range is shown. The quality of this data is typical for all of the samples investigated. As can be seen there are multiple frequencies, some of which are harmonics of fundamental frequencies, and some are fundamental frequencies plus or minus another fundamental frequency due to magnetic interactions, i.e., the B field seen by electrons on one extremal area orbit is modulated due to oscillations of electrons on other extremal area orbits. In Fig. 1 the peaks in the Fourier transform at 12 and 18 kT are the second and third harmonics of the peak at 6 kT. Each reported fundamental frequency given below has been checked to assure it is not either of the nonfundamental frequencies described above.

III. RESULTS AND DISCUSSION

The results of the angular dependent measurements on the three $LaMIn_5$ compounds are shown in Figs. 2–4 for $M=Rh, Ir, \text{ and } Co$, respectively. The first observation is that the number of frequencies observed increases as one proceeds from $3d(Co)$ to $4d(Rh)$ and then to $5d(Ir)$ contributions to the conduction bands. This result indicates that there are different interactions between the d band electrons and the free-electron s band states depending on increasing angular momentum of the d electrons. The first results we obtained were for $LaRhIn_5$ with the measured frequencies as a function of angle shown in Fig. 2. The consequences of the very low frequency (7 T) that we have reported on previously⁹ are not discussed in this paper. Overall these results for $LaRhIn_5$ are consistent with those obtained by Shishido *et al.*¹⁰ In the Rh case band calculations can be done accurately, displaying remarkable agreement with both the experimental results reported in Ref. 10 as pointed out in Ref. 5. In Figs. 3 and 4 the results of the first measurements of the dHvA effect on

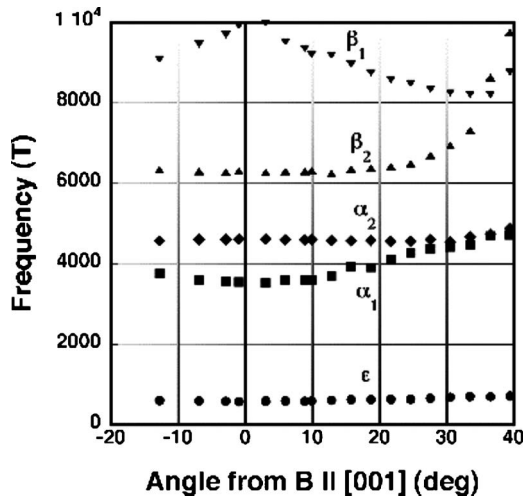


FIG. 2. dHvA frequencies vs angle between [001] and the field direction for LaRhIn₅.

LaIrIn₅ and LaCoIn₅ are shown. In these figures the solid lines are fits to $f=f_0/\cos(\theta)$, where f_0 is the frequency at $\theta=0$, over limited angular ranges. The frequencies that are quasicylindrical all arise from extremal area orbits on the band 15-electron part of the FS, normally denoted as α_1 , α_2 , and α_3 . The α_1 , α_2 , and α_3 frequencies all arise from a single piece of FS, electron band 15 (see Fig. 5). Were this piece to be perfectly cylindrical there would be only one a orbit that would have the $1/\cos(q)$ angular dependence. Because this piece of FS is actually an undulating cylinder, as shown in Fig. 5, there are three a orbits corresponding to maximum and minimum areas along the axis of the cylinder. Larger undulation leads to a larger frequency separation for the three orbits. We note that, while cylindrical-like orbits are seen in LaCoIn₅ and LaIrIn₅, they are more separated than those seen in CeCoIn₅ (Ref. 6) and CeIrIn₅ (Ref. 8) indicat-

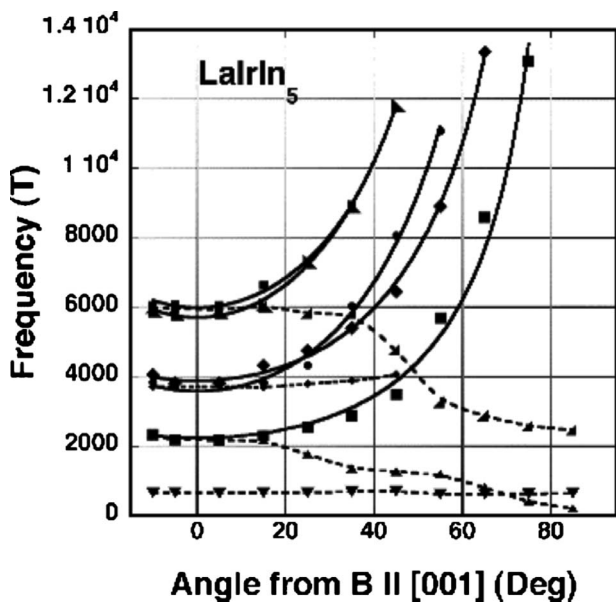


FIG. 3. dHvA frequencies vs angle between [001] and the field direction for LaIrIn₅.

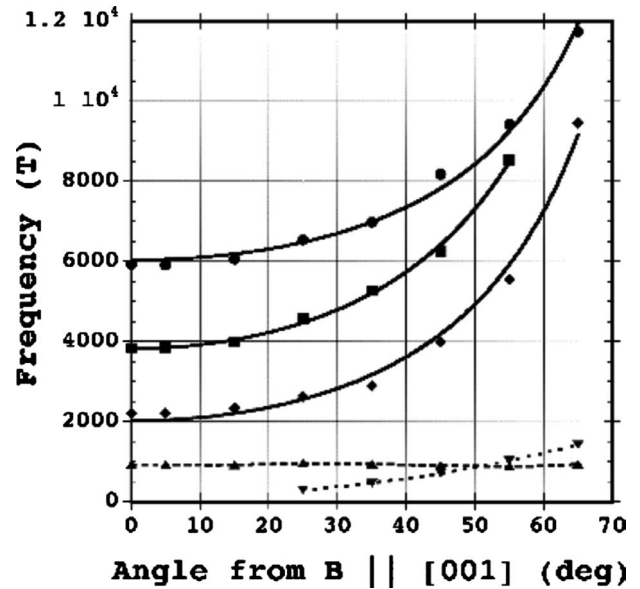


FIG. 4. dHvA frequencies vs angle between [001] and the field direction for LaCoIn₅.

ing that the f electrons also cause this piece of the FS to become more cylindrical. The observation to be made here is that the overall interaction of the d levels of the M in the CeMIn₅ compounds is significant and the Ce f levels are not the only electron states involved in determining the quasicylindrical nature of the FS shapes. They do, however, cause increases in effective mass in each case. As was previously shown, the partially occupied f bands for the Co and Ir Ce-based compounds cause increases in the FS dimensions while the Rh-based Ce and La FSs remain nearly identical.¹¹

As stated above comparing the results of measurements on LaRhIn₅ and CeRhIn₅ show that, due to the nearly complete f electron localization in the Ce compound, the Fermi surface of the two compounds are almost the same. We now compare the current results for the three α orbits in the other two La compounds, Ir and Co, to their Ce-based counterparts. In the case of LaIrIn₅ for $\theta=0$ the three α frequencies are approximately 2, 4, and 6 kT. In CeIrIn₅ they are 4.2, 4.5, and 5.6 kT.⁸ For LaCoIn₅ the frequencies are again approximately 2, 4, and 6 kT whereas in CeCoIn₅ they are separated by less than 1 kT.⁶ This shows that the undulation

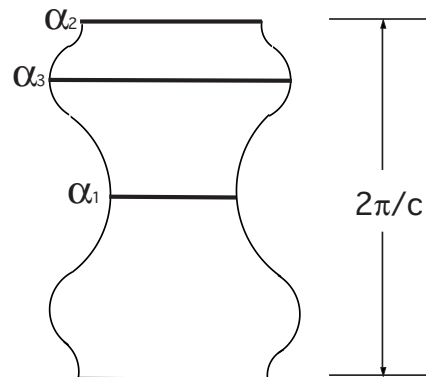


FIG. 5. The α orbits in these materials arise from the maxima and minima of a Fermi surface shaped like an undulating cylinder.

in the cylindrical nature of the Fermi surface due to the band 15 electrons has been significantly reduced by the interactions with the $4f$ electrons leading to a more layered structure, 2D like, for this electron band in the two superconducting compounds. We also point out that the least undulation occurs in the Co compound which has the highest T_c .

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