

## **dHvA measurements on LaSb<sub>2</sub>**

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### **Abstract**

In this paper the results of de Haas – van Alphen (dHvA) measurements in high magnetic fields on LaSb<sub>2</sub> are reported and compared to the results of energy band calculations for the known structure of LaSb<sub>2</sub>. Three dHvA frequencies are observed that are associated with three of the Fermi surface pieces from the energy band calculation. There is agreement between these measured Fermi surface areas and three of the calculated areas, but other frequencies found in the band calculation are not observed experimentally. The effect that the result of these Fermi surface measurements have on the large linear magnetoresistance found in LaSb<sub>2</sub> is discussed.

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## Introduction

Materials that exhibit a large magnetoresistance (MR) have received a great deal of attention in the past decade because of their potential use as magnetic field sensors in read-write heads. Magnetic materials are often employed, since suppressing magnetic fluctuations in a magnetic conductor can decrease carrier scattering and thus result in a large MR. There are also several notable examples of nonmagnetic materials where large MRs result from the Coulomb interactions between carriers. Most intriguing are materials where no suitable mechanism is known for the large MR. This last group includes AgSe and the rare earth di-antimonides where large linear field dependent resistivities have been measured over wide ranges of magnetic field. Included in this latter group is the metal LaSb<sub>2</sub> which exhibits a large linear magnetoresistance (MR) showing no sign of saturation up to fields of 45 T<sup>1</sup>. The standard semiclassical theory<sup>2</sup> of MR that includes band structure effects predicts a resistance that increases quadratically with magnetic field, unless there are open Fermi surface (FS) orbits, in which case the MR eventually saturates at high field. Theories that have been successful in predicting a linear-like MR over small ranges of magnetic field rely on either magnetic breakdown effects in charge density wave (CDW) materials<sup>3</sup> or high field quantization effects<sup>4</sup>. LaSb<sub>2</sub>, and the other rare earth di-antimonides, have anisotropic 2 dimensional-like crystal structures similar to the classic CDW compounds NbSe<sub>2</sub> and TaSe<sub>2</sub>, both of which display large linear MRs. However, no CDW ground state has been established for LaSb<sub>2</sub>.

In this paper we report the results of de Haas – van Alphen (dHvA) measurements on LaSb<sub>2</sub> and compare them with the results of our energy band calculations for the

known structure of  $\text{LaSb}_2$ . We find very good agreement between the measured FS areas and three of the calculated areas, indicating that, if a CDW ordering occurs in this material, it affects neither the pieces of the Fermi surface near the zone center nor those parts near the zone boundaries at the S point. Furthermore, those pieces of the Fermi surface that a naive nesting picture would be predicted to be involved in a CDW ordering are not observed in our measurements. This indicates that a CDW ground state may appear in  $\text{LaSb}_2$ .

### Experimental Method

$\text{LaSb}_2$  is a member of the  $\text{RSb}_2$  ( $R=\text{La-Nd, Sm}$ ) family of compounds that all form in the orthorhombic  $\text{SmSb}_2$  structure<sup>5</sup>.  $\text{LaSb}_2$  is comprised of alternating La/Sb layers and two-dimensional rectangular sheets of Sb atoms stacked along the  $c$ -axis. Similar structural characteristics give rise to the anisotropic physical properties observed in all the compounds in the  $\text{RSb}_2$  series<sup>6</sup>. Since  $\text{LaSb}_2$  is non-magnetic, its low-temperature properties are not complicated by magnetic phase transitions which occur in the other members of this series<sup>6</sup>. Single crystals of  $\text{LaSb}_2$  were grown from high purity La and Sb by the metallic flux method<sup>7</sup>. The structure and quality of these crystals has been reported previously<sup>1</sup>.

Measurements of the de Haas – van Alphen effect were made in slowly swept magnetic fields between 20 and 45 T at the National High Magnetic Field Laboratory, Tallahassee, FL. The sample torque magnetization was measured using a rotatable cantilever system, and data was taken as a function of angle with respect to the direction of the applied magnetic field in  $5^\circ$  steps over a  $90^\circ$  angular range in the  $a$ - $c$  plane. In addition, effective mass measurements were made near  $\text{B}_{||}[001]$  from the temperature dependence of the dHvA signals.

## Results and Discussion

In Fig. 1 a typical data set is shown with a Fourier transform of the data shown in the inset. Because of the low frequencies observed giving rise to a small number of oscillations in the field range of measurement, we fit the data set to a sum of three frequencies using the Lifshitz – Kosevitch (LK) equation<sup>8</sup>. The result of the fit is shown as a dotted line in Fig. 1. As can be seen from the overall fit constant frequencies are observed up to 45 T with no changes in the various Fermi surface sizes or shapes. The two highest frequencies obtained from the fit are in agreement ( $\pm 2\%$ ) with those obtained from the Fourier analysis. However, the lowest frequency differs in the fit considerably from that obtained from the Fourier transform. By inspection of the data it can be seen that there are at most 1.5 oscillations of this low frequency in the field range of 20 to 45 T and therefore the Fourier transform is less reliable. In the analysis that follows we have used the frequencies obtained from the LK fits to obtain the frequencies rather than the peaks in the Fourier transform spectrum.

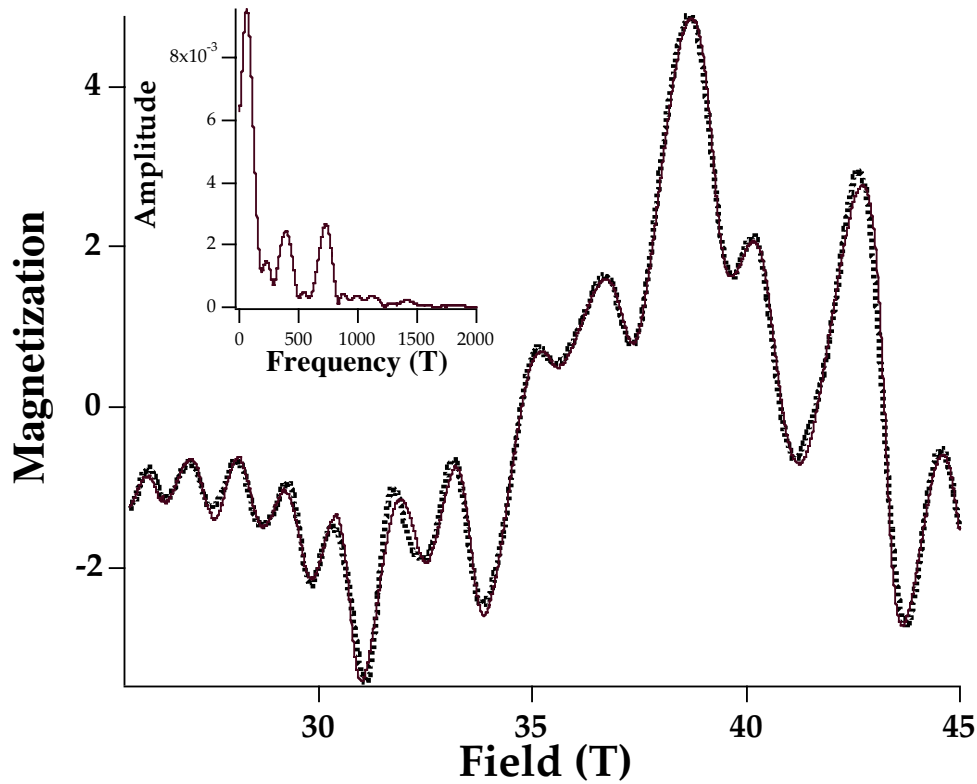


Figure 1. Magnetization as a function of field, solid line, of LaSb<sub>2</sub>. The dotted line is a fit to the data for the three frequencies shown in the Fourier transform of the data in the inset.

In Fig.2 the angular dependences of the three frequencies found in Fig.1 are shown. As noted above, the two higher frequencies are essentially the same whether they are taken from the LK fit or the Fourier spectrum. From the angular dependence of these three frequencies it can be seen that because of the angular behavior compared to a  $1/\cos(\theta)$  dependence on angle of the two higher observed frequency signals that would be expected if the FS were truly two dimensional, they arise from Fermi surface sheets that appear to be connected along the  $\Gamma - \Gamma$  direction in the BZ, but not perfectly cylindrical. The small frequency is nearly angle independent and arises from a nearly spherical small sheet. While the value of the small frequency differs

between the two methods of fit, both methods show that the frequency is nearly independent of angle.

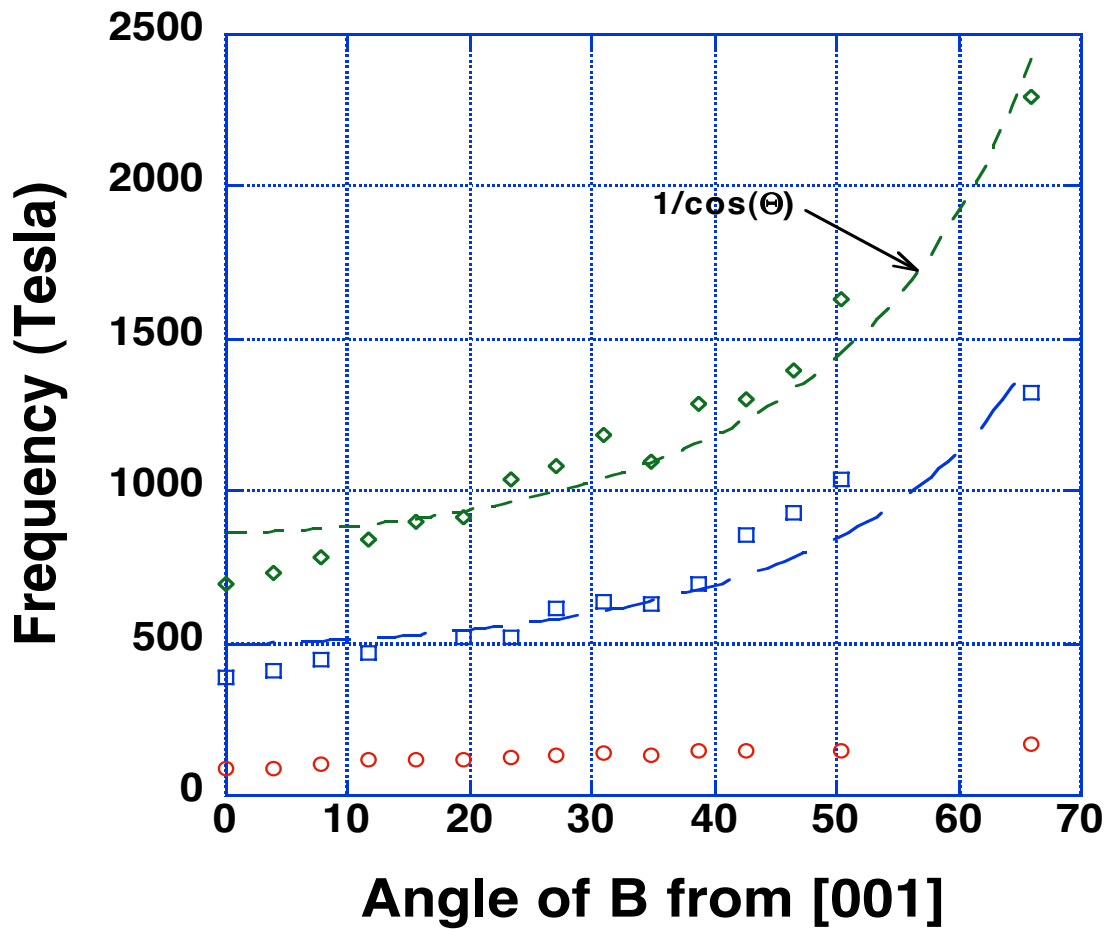


Figure 2. Angular dependence of three dHvA frequencies for rotations from zero degrees at B|| [001] toward B||[100].

From the temperature dependence of the amplitude of the Fourier transform of the dHvA signals 15° from the [001] we find the effective masses of the two higher

frequencies to be  $m^* = 0.22$  for the 400 T signal and  $m^* = 0.17$  for the 700 T signal. All of the signals were clearly observable up to 15 K indicative of the light masses. No mass of the lowest frequency could be obtained.

We also performed energy band calculations as part of the overall investigation of the electrical properties of  $\text{LaSb}_2$ .<sup>9</sup> The band calculations used the WIEN2K software package<sup>10</sup>, which implements full potential all-electron correlation potentials of Perdew et al<sup>11</sup>, and the spin-orbit interaction was included perturbatively. The La and Sb muffin tin radii were set at 2.85 and 2.65 a.u. respectively. A plane-wave cutoff of  $5 \text{ au}^{-1}$  was employed, resulting in a total of 649 plane waves. Semi-core states for La s, p and Sb s, d states were included with 216 local orbitals, and a total of 144 k-pts were kept in the irreducible Brillouin zone. After the calculation had converged, a much finer grid of  $81 \times 81 \times 11$  points was employed to calculate the FS. Cross-sections of the overall FS obtained from these calculations in the  $\Gamma Y_1 Y_2$  plane (Zone Center) are shown in Figure 3a for a single zone and in Figure 3b in a repeated zone scheme. We associate the three observed frequencies for  $B \parallel [001]$  with the three FS orbits marked  $F_1$ ,  $F_2$ , and  $F_3$ . For the smallest frequency,  $F_1$ , the band calculation is the least accurate because the grid would have to be much finer to obtain accurate results. Therefore, it is not clear which band gives rise to  $F_1$ , but it should be one corresponding to the small orbits near the S point, all of which are ellipsoidal in shape. This is consistent with the angular dependence presented in Fig.2. The orbits marked  $F_2$  and  $F_3$  are associated with sheets that are rather two-dimensional and show rather small variation in cross-section along the  $[001]$  direction, which is consistent with the angular dependence seen for the two higher frequency components.

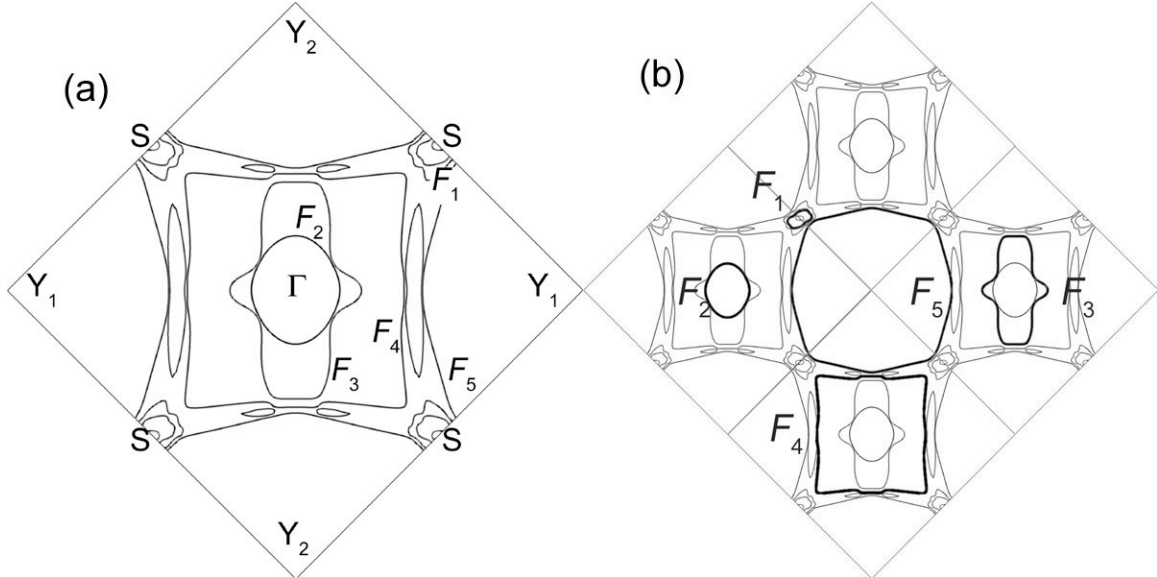


Figure 3. (a) Fermi surface contours in the center of the zone with orbits indicated by F1–F5; (b) extended zone with each of the closed orbits for  $B \parallel [001]$  highlighted.

In the Table we give the values of the calculated and measured dHvA frequencies in  $\text{LaSb}_2$  for the applied field parallel to the  $c$  axis, or lying in the layered planes. As can be seen the agreement for  $F_1$ ,  $F_2$ , and  $F_3$  is very good. There are two other large pieces of FS in the  $\Gamma Y_1 Y_2$  plane arising out of the energy band calculations that are not observed in the measurements. They are labeled  $F_4$  and  $F_5$  in Figure 3. Due to the fact that the measurements were made to 45 T, and the effective mass of all of the carriers is not large, the two higher frequencies easily should have been observed from the cantilever measurements.

Table

	<b>Band Calculations</b>	<b>Experiment</b>
<b>F<sub>1</sub> (tesla)</b>	86	102
<b>F<sub>2</sub> (tesla)</b>	386	393
<b>F<sub>3</sub> (tesla)</b>	737	729
<b>F<sub>4</sub> (tes;a)</b>	1900	Not Observed
<b>F<sub>5</sub> (tesla)</b>	3800	Not Observed

There are at least two possible reasons that the frequencies  $F_4$  and  $F_5$  are not observed experimentally. The first is that magnetic breakdown could be occurring at points on both of the orbits near the  $\Gamma Y_1$  or  $\Gamma Y_2$  directions. This possibility would lead to a series of orbits, some closed and some open that are not distinct or may not be observed. Second, if a CDW exists at low temperatures, the nesting of the wave-vector is most prominent along the sheets of the Fermi surface associated with  $F_4$  and associated  $F_5$ . We would expect those parts to be removed by the formation of the CDW gap, or if the CDW formed only along one direction, to become part of an open orbit. In distinction, the parts of the FS located near the high symmetry points in the zone such as  $F_2$  and  $F_3$  centered at  $\Gamma$ , would be unaffected by the presence of a CDW. In Ref. 9 the nesting between branches of these parts of the FS that would lead to CDW formation is discussed in detail.

In conclusion, we show that two sheets of the FS of  $\text{LaSb}_2$  extend along the  $\Gamma X$  direction leading to open orbits when the field is applied perpendicular to the  $[001]$  axis.

This fact is contrary to the accepted MR arising from metals with open orbits<sup>Error! Bookmark not defined.</sup> and fails to account for the un-saturating linear MR found in LaSb<sub>2</sub> for the field in the plane of the sample. However, the linear nature of the measured MR is not explained by open orbits because the MR should be quadratic in field for breakdown to be the entire cause. The lack of observation of the network of orbits could be the result of a CDW forming at low temperatures that has yet to be observed directly.

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