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Shubnikov–de Haas and High-Field Magnetoresistance Effects in the A15 Compound Nb_3Sb*

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High-field magnetoresistance and Shubnikov–de Haas (SdH) effects were studied in the A15 compound Nb_3Sb in fields up to 215 kG. A change in the field dependence of the magnetoresistance for certain field directions above about 150 kG appears to signal the onset of magnetic breakdown. Five sets of SdH frequencies were observed, four of them closely corresponding to de Haas–van Alphen (dHvA) frequencies observed by Arko et al. *The fifth frequency had an extremely large amplitude, about 20% of the backgroud magnetoresistance, and it is suggested that this also is due to magnetic breakdown. The results are compared with the ab initio band calculations of van Kessel et al., which can explain many of the observed features of the dHvA and SdH frequencies.*

The A15 compounds have been of great interest in recent years for several reasons. They include many of the highest T_c superconductors, such as Nb_3Ge and Nb_3Sn. They also exhibit a number of interesting physical properties, such as structural distortions and an anomalous electrical resistance. The interplay among the electrons, phonons, and structure is clearly of interest and the band structure ultimately will be an essential part of the detailed understanding of these materials (see Ref. 1 for a recent review).

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Arko *et al.*[2] recently have reviewed the status of Fermi surface measurements in the A15 compounds and have compared the results to the available band structure calculations. The experimental measurements are limited by the difficulty in preparing high-quality single crystals and by the necessity to perform the experiments above H_{c2} ($\geq$200 kG in Nb_3Sn and V_3Si). However, for Nb_3Sb the situation is much more favorable, since H_{c2} is low and excellent single crystals can be grown. This has led to detailed de Haas-van Alphen (dHvA) measurements in this compound as reported by Arko *et al.*[2,3]

At the time of the first dHvA paper[3] there had been no band structure calculations on Nb_3Sb. It was possible only to construct a partial Fermi surface model. This model included two small pockets and a sheet partially cylindrical in shape whose connectivity was open to question. These results led to the present measurements, whose purpose was to probe the topology of the Fermi surface of Nb_3Sb, and to search for any Shubnikov–de Haas (SdH) oscillations which might supplement the dHvA frequencies of Arko *et al.*

The single-crystal sample of Nb_3Sb used in this study was grown by an iodine vapor-transport technique. The current axis of the sample was $\langle 110 \rangle$, its dimensions were about $1 \times 1 \times 3\ \mathrm{mm}^3$, and its resistance ratio $\rho(295\ \mathrm{K})/\rho(4.2\ \mathrm{K})$ was 90.

Because of the extreme difficulty in attaching electrical leads to Nb_3Sb, a brief description of the technique that was successful will be given here. A variety of solders, silver paint, and conductive epoxy all proved to be failures as electrical contacts. The successful technique involved first scraping the surface of the crystal with a sharp instument to remove a thin layer of insulating material, perhaps an oxide. Then, four extremely small dots of a liquid metal alloy (Viking Metal) were placed on the crystal for the current and potential contacts. After insertion of four tinned copper wires into the four dots, each wire/dot combination was painted with GE 7031 varnish for mechanical stability. Even this procedure was successful in only about 50% of the attempts.

The measurements were made at 4.2 K in a 3-cm-bore Bitter solenoid at the National Magnet Laboratory. The maximum field employed was about 215 kG.

Figure 1 shows a transverse magnetoresistance rotation plot at $B =$ 211 kG. There are several points to be made concerning this figure. The first is that the magnetoresistance plot is not perfectly symmetric about [001] as it should be. The most likely explanation for this is that the extremely small size of the sample, especially the small length-to-width ratio, made both the positioning of the sample on the holder and the positioning of the potential leads on the sample much less than ideal. These effects have led to similar

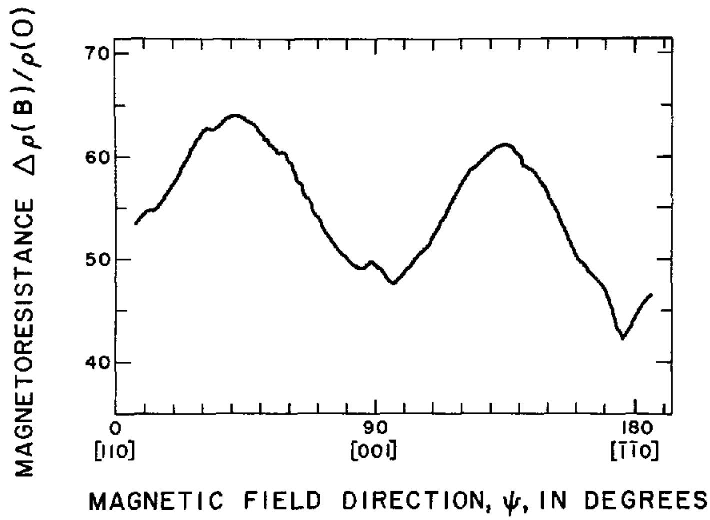

Fig. 1. Transverse magnetoresistance of Nb_3Sb with current density J‖⟨110⟩ at a field of 211 kG and at 4.2 K.

asymmetries in other compounds we have investigated (see, e.g., Refs. 4). Second, the maximum value of the magnetoresistance is 64, which would imply a value of about 8 for $\langle\omega_c\tau\rangle_{av}$, where ω_c is the cyclotron frequency and τ the relaxation time. This crude estimate, based on a simple two-band model,[5] indicates that the sample purity is at least high enough for the

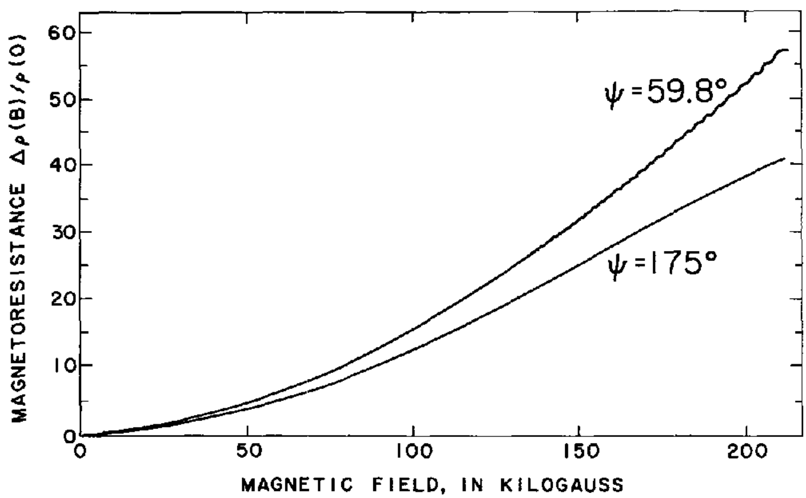

Fig. 2. Field dependence of the magnetoresistance for two angles shown in Fig. 1.

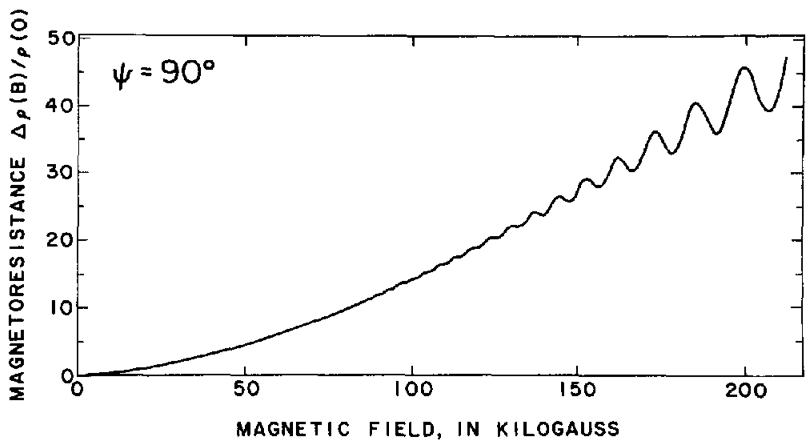

Fig. 3. Example of large-amplitude SdH oscillations for $\psi = 90°$ in Fig. 1.

high-field region, $\langle \omega_c \tau \rangle_{av} > 1$, to be approached. Third, the oscillations on either side of the maximum at about 45° are SdH oscillations; their amplitude is seen clearly in the top curve of Fig. 2, which is a field sweep at $\psi = 59.8°$. Each oscillation in Fig. 1 is the result of the argument of the oscillatory magnetoresistance term $\alpha \equiv 2\pi F/H$ changing by 2π; that is, the SdH frequency F is a function of ψ. Fourth, there is a fairly sharp dip in Fig. 1 at $\psi = 175°$. The corresponding field dependence at this position is seen in the lower curve of Fig. 2. In contrast to the upper curve, for which $\Delta\rho/\rho \propto B^2$, the magnetoresistance at the dip is beginning to saturate above about 150 kG. This has implications either for the topology of the Fermi surface or for a change in the state of compensation. Fifth, in Fig. 1 there is a rather "unnatural" maximum in $\Delta\rho/\rho$ near $\psi = 90°$. It is odd because it occurs where the rest of the data would suggest a minimum,* and because it lacks symmetry about its local maximum. The field dependence of the magnetoresistance was investigated carefully in the vicinity of this peak; an example for $\psi = 90°$ is shown in Fig. 3. It is seen that a large-amplitude SdH oscillation is present. The size of this oscillation is quite suggestive of a magnetic breakdown process in which an open orbit is being successively turned on and off as various Ladau levels sweep through a particular sheet of Fermi surface. In fact, the maxima of these oscillations appear to follow an approximate quadratic field dependence, while the minima exhibit a tendency toward saturation.

Figure 4 gives a summary of all the SdH frequencies observed along with a composite of the dHvA frequencies seen by Arko *et al.*[2] The dHvA

*Another example of this behavior was seen in AuSn.[6]

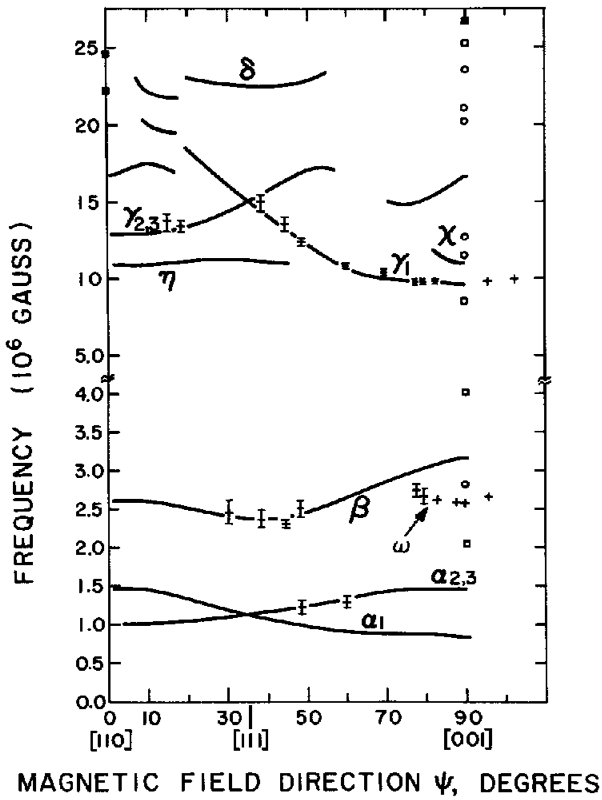

Fig. 4. Summary of SdH frequencies observed in the (110) plane (plus signs). The solid lines are curves representing dHvA frequencies measured by Arko *et al.*[2] The circles and squares are additional dHvA frequencies measured by Arko *et al.* It should be noted that the data points for $\psi > 90°$ are also for fields in the (110) plane.

frequencies are shown by the solid lines in the (110) plane and by the circles and squares at [110] and [001]. The SdH frequencies observed correspond to the $\alpha_{2,3}$, β, γ (including beats), and $\gamma_{2,3}$. Of special interest is a new frequency, denoted here as ω, apparently not observed in the dHvA work. The ω frequency is observed only near [001] and is the oscillation for which the evidence suggests (see Fig. 3) that magnetic breakdown may be the cause.

The results presented above lead to two significant questions. These are: (1) Does the tendency for $\Delta\rho/\rho$ to saturate for $\mathbf{B}\|\langle 110\rangle$ and $\langle 001\rangle$ correspond to a multiply-connected Fermi surface or a loss of compensation due to magnetic breakdown? (2) What is the origin of the newly observed ω frequency? To discuss question 1, the state of compensation of Nb_3Sb must be discussed. Since the A15 structure contains two molecular units (i.e., six atoms), the total electron count per (simple cubic) primitive cell is even. This indicates that Nb_3Sb should be a *compensated* metal with equal electron and hole Fermi surface volumes. According to the theory of high-field magnetoresistance effects,[5] this means that the magnetoresistance should saturate only if there is an open orbit perpendicular to the field and to the current, or the field is in a high-symmetry direction for which the normal state of compensation is destroyed; this latter situation is called *geometric discompensation* when the field is in a *singular field* direction. Under normal circumstances geometric discompensation can occur only in a compensated metal which has a multiply connected Fermi surface; if all the sheets are closed surfaces it is impossible for the compensated state to be destroyed. The field dependence at $\mathbf{B}\|\langle 110\rangle$ and $\mathbf{B}\|\langle 001\rangle$ suggests that the tendency toward saturation is in fact due to a change in the state of compensation from compensated to uncompensated in fields above about 150 kG, a change due to magnetic breakdown. The reason for this statement is that experience on many compounds[7] has shown that if saturation is to occur for reasons of open orbits, the magnetoresistance generally does not change from upward curvature to downward curvature above some characteristic field—it is always curving downward, assuming the high-field condition is satisfied.

There are two other reasons to suspect magnetic breakdown on the Fermi surface of Nb_3Sb. One already mentioned is the large amplitude of the SdH oscillations (~20%) seen in Fig. 3 for **B** near $\langle 001\rangle$. The second is that ab initio band calculations of van Kessel *et al.*[8] have shown that there are several pieces of Fermi surface which are degenerate, or very nearly degenerate, along certain high-symmetry lines in the Brillouin zone. The Fermi surface calculated by van Kessel *et al.* exists in bands 18–24, and consists entirely of closed surfaces centered on the M and R points of the zone. An inkling of the nature of these surfaces and the above-mentioned degeneracies can be seen in the cross sections of van Kessel *et al.*,[8] shown in Fig. 5. These sections are in an RMX or (001) plane in the Brillouin zone.

The second question raised above, that of the origin of the ω frequency, also can be discussed with reference to Fig. 5. Our ω frequency, observed only out to about 12° away from $\langle 001\rangle$, is not far from that expected for band 19, and is also not far from a frequency observed by Arko *et al.*, shown as an open circle just above our ω frequency. It is not clear whether our ω frequency is in fact the same as the dHvA frequency, or whether it corresponds to band 19. The dHvA work appears to have seen two of the expected branches of band 19 but not the third, and again this may be due to magnetic breakdown of the orbits passing close to the RM near-degeneracy

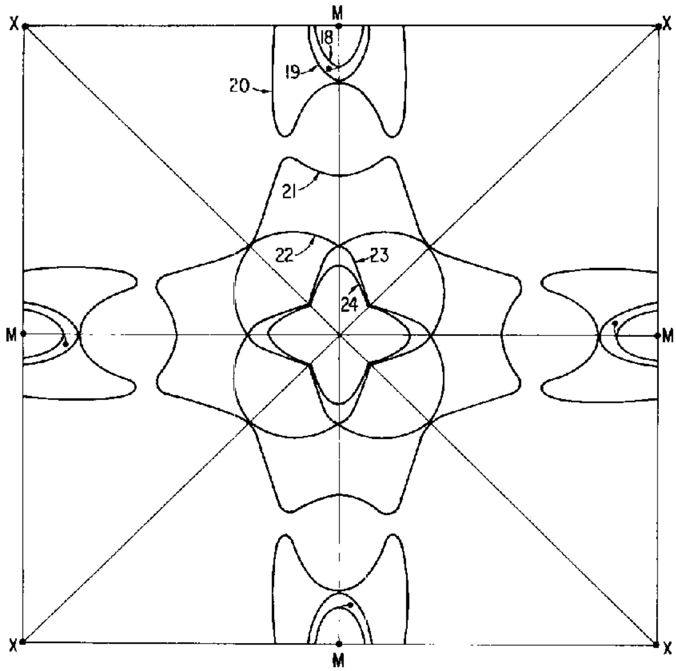

Fig. 5. Fermi surface cross sections in the (100) plane taken from the ab initio band calculation of van Kessel *et al.*[8]

of bands 19 and 20. The upward curvature of the ω frequency as **B** moves away from ⟨001⟩ appears to be real and it is not obvious that the band structure results lead to this behavior. There is another problem in associating the ω frequency with band 19. This is that, as mentioned earlier, generally when such large-amplitude SdH oscillations exist they are due to a magnetic breakdown process in which an open orbit is being successively turned on and off as the Landau levels of a (typically small) piece of Fermi surface pass through that surface. This small sheet then forms a "link" between two sheets of Fermi surface in different zones so that the open orbit is switched on and off with the SdH frequency of the small linking sheet. A well-defined example of this is found in Be, where breakdown between the "coronets" and "cigars" leads to open orbits which are modulated by the cigar SdH frequency.[9] The problem then is that if the ω frequency is associated with band 19, there is no mechanism by which breakdown can lead to open orbits via another sheet of Fermi surface. One can imagine, however, breakdown between the "stumps" of band 20 (hole) and band 21 (electron) shown in Fig. 5. This could cause egg-shaped cyclotron orbits whose area corresponds to the ω frequency. This situation has been considered by Mueller,[10] who finds it altogether feasible and also points out that such orbits would increase in frequency as **B** is tipped away from [001], as observed.

The above hypothesis also is consistent with the overall magnetoresistance behavior. That is, for $\mathbf{B}\parallel[\bar{1}\bar{1}0]$ (Fig. 1) there is a tendency toward saturation which could be caused by a loss of compensation due to breakdown, but no ⟨110⟩ open orbits or large SdH oscillations which would be consistent with Fig. 5. However, when $\mathbf{B}\parallel[001]$, open orbits in ⟨001⟩ directions could be induced by magnetic breakdown, with large SdH oscillations, as proposed above.

In conclusion, with the exception of the origins of the magnetic breakdown as explained above, the adjusted band calculation of van Kessel *et al.*[11] gives a good basis for correlating most of the dHvA and SdH frequencies with Fermi surfaces in bands 18–24. The present results in open orbits, magnetic breakdown, and the observation of the ω frequency all suggest that additional theoretical efforts may be worthwhile to refine certain aspects of the Fermi surface of Nb_3Sb.

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