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Oscillatory Magnetoresistance in 4*Hb*-TaS_2 *†

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Oscillations with frequencies in the range 0.04 to 3 MG have been observed in the magnetoresistance of 4*Hb*-TaS_2. The low frequencies observed are consistent with sections of Fermi surface resulting from band folding due to the charge-density-wave superlattice, and the angular dependence of the frequencies suggests cylindrically shaped pieces of the Fermi surface. For particular lead geometries the oscillatory portion of the experimental voltage displays an unusual current dependence in that it scales linearly with $|\bar{I}|$ but does not depend on the direction of $\bar{I}$.

Shubnikov–de Haas oscillations have been observed in the magnetoresistance and Hall resistance of the layer compound 4*Hb*-TaS_2 in the temperature range 0.62–4.2 K and in magnetic fields up to 220 kG. Five frequencies in the range 0.04 to 3 MG have been identified and correspond to sections of Fermi surface containing from 10^{-4} to 10^{-2} electrons per atom. These correspond to very small cross sections and are consistent with a Fermi surface broken up by the charge-density-wave (CDW) superlattice. The angular dependence for field orientations between parallel and perpendicular is consistent with that expected for cylindrically shaped sections of Fermi surface. Measurements for particular lead and crystal geometries show an unusual and novel behavior of the oscillations in that they appear as terms which depend only on the magnitude of the current $|\bar{I}|$ and not on the direction of $\bar{I}$.

The crystals were grown by iodine vapor transport in a gradient of 720–700°C for four weeks following the method of Wattamaniuk, Tidman, and Frindt.[1] Polyhedral crystals several millimeters in diameter have been mounted with the current flow parallel to the layers in the [10$\bar{1}$0] direction. Because of the geometrical variation of the crystal cross section and the large anisotropy of conductivity the current may be quite non-

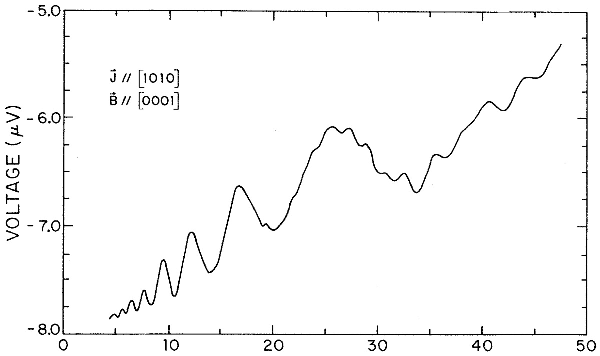

FIG. 1. Shubnikov–de Haas oscillations observed in $4Hb$-TaS_2 at 0.62 K for magnetic field perpendicular to the layers.

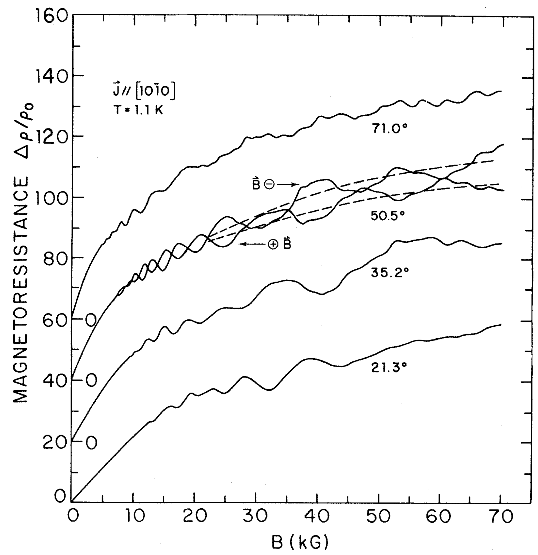

FIG. 2. Field dependence of magnetoresistance for four field orientations in plane perpendicular to the basal plane. Curve for $\theta = 50.5°$ shows sweeps for $\pm\overline{B}$. Dashed curves show asymmetry in the dc magnetoresistance due to Hall component.

uniform. On the as-grown specimens, potential leads have been placed on the finely stepped side surfaces or on a single basal-plane facet forming the top surface. Four-lead measurements have also been made on thin cleaved sections where the geometry favors more uniform current flow. Copper contacts attached with silver paint or indium have been used.

The field dependence of resistance for a magnetic field perpendicular to the layers at a temperature of 0.62 K is shown in Fig. 1. The oscillations are periodic in $1/\overline{B}$ and are observed over most of the angular range of $\overline{B}$ between $B_\perp$ and $B_\parallel$ as shown in Fig. 2. The detailed angular dependence of two of the frequencies is shown in Fig. 3 and both follow a dependence of the form $\omega_\perp/\sin\theta$, as represented by the solid lines, where $\omega_\perp$ is the frequency observed for field perpendicular to the layers. The oscillatory structure appears to scale with orientation as $1/\sin\theta$, although the higher frequencies are difficult to track since at lower angles of $\overline{B}$ only a few periods are observed before reaching the maximum field of 220 kG. At field angles near perpendicular, the individual high frequencies can be separated and values of 0.9, 1.2, and 3 MG have been observed in addition to those shown in Fig. 3. For $\overline{B}$ less than 10° from the parallel orientation, no oscillations are observed although structure develops in the high-field magnetoresistance rotation curves. Sharp peaks suggesting the presence of open orbits on the Fermi surface parallel to the basal plane are observed, but the field dependence of magnetoresistance does not approach the expected quadratic dependence in the available field of 220 kG. The field dependence of the Hall resistivity shows changes in slope which suggest the presence of magnetic breakdown.

Magnetoresistance measurements on thick as-grown specimens display an anomalous oscillatory voltage component which does not reverse with

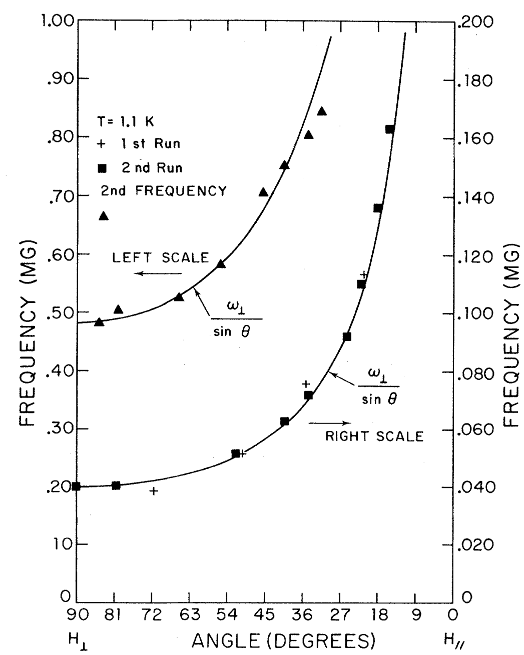

FIG. 3 Angular dependence of two frequencies measured for field orientations in a plane perpendicular to the basal plane.

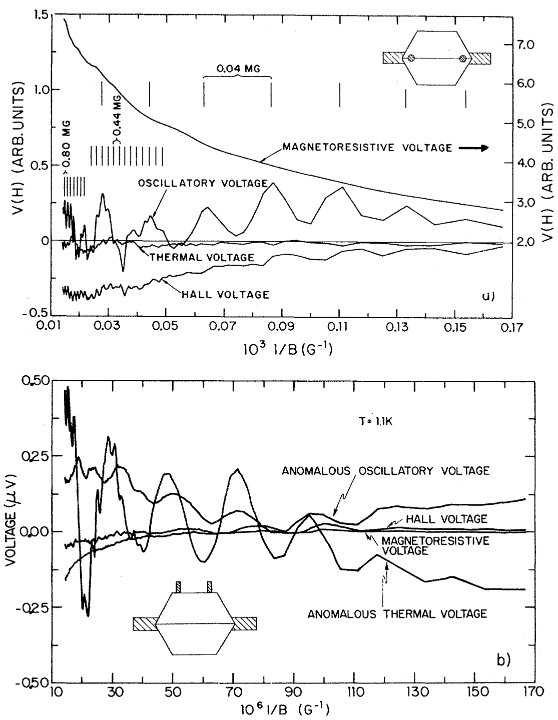

FIG. 4. Separated components of the voltage obtained by appropriate summing of the four terms $(1) = V(+\overline{I}, +\overline{B})$, $(2) = V(-\overline{I}, +\overline{B})$, $(3) = V(+\overline{I}, -\overline{B})$, and $(4) = V(-\overline{I}, -\overline{B})$ plotted against $1/\overline{B}$. Lead configurations are shown in insets. (a) Dominant oscillations occur in the term (1) + (2) − (3) − (4) which is odd in $\overline{B}$ and dependent only on $|\overline{I}|$. (b) Dominant oscillations occur in the term (1) + (2) + (3) + (4) which is even in $\overline{I}$ and $\overline{B}$.

current direction. Depending on specific lead geometries, this anomalous oscillatory component can show either an odd or an even dependence on the field direction. In both cases the amplitude of the oscillatory component scales linearly with the magnitude of $|\overline{I}|$. These effects are illustrated in the four-term analysis of Fig. 4. These curves show the conventional terms obtained from a combination of measurements obtained for $\pm\overline{I}$ and $\pm\overline{B}$. The major amplitudes of oscillation occur in the terms that are even in $\overline{I}$ as opposed to the usual magnetoresistance and Hall terms which are odd in $\overline{I}$. Figures 4(a) and 2 correspond to the lead configuration with leads on the finely stepped side surfaces of the crystal in the same plane as the current leads. In this case the dominant oscillations are even in $\overline{I}$ and odd in $\overline{B}$. Figure 4(b) corresponds to the lead configuration with the leads on the top surface of the crystal. In this case the dominant oscillations are even in $\overline{I}$ and even in $\overline{B}$ as well. In both lead configurations the dc magnetoresistance becomes fairly smooth in the four-term analysis.

If the three-dimensional crystals are cleaved into thin specimens so that a uniform cross-section and a more standard lead configuration and current distribution can be achieved, the oscillations revert to an odd dependence on current and appear in the magnetoresistance and Hall terms. If in this same thin crystal one rearranges the leads so that the current flows only in the first half of the specimen and the potential is measured over the second half using separate leads, the same anomalous oscillatory voltage is observed as in the three-dimensional case illustrated in Fig. 4(b). The measured frequencies are the same for both the anomalous and normal oscillatory terms.

These results tend to confirm the conclusion that a voltage not associated with the usual dissipative terms of the resistivity tensor is being observed. An unusual voltage of this type has recently been reported by Egorov[2] for measurements on a single crystal of beryllium; however, he reports that the emf shows a nonlinear current dependence. He has suggested a possible mechanism based on a nonequilibrium effect produced in the electron gas by open orbits resulting from magnetic breakdown. Our experiments certainly give preliminary evidence of open orbits and magnetic breakdown in $4Hb$-TaS_2, but the anomalous terms in the oscillatory component show no dependence on angle except for the frequency variation reported above.

The low frequencies observed in the present experiment indicate the existence of sections of Fermi surface 10–100 times smaller in cross section than expected from the band-structure calculations for either the $2H$ polytype or the $1T$ polytype without the CDW superlattice. The Fermi-surface cross sections constructed by Wilson, Di Salvo, and Mahajan[3] from the augmented-plane-wave band-structure calculation of Mattheiss[4] for $2H$-TaS_2 give frequencies in the range 30 to 150 MG. In the CDW state one must recalculate the band structure in the reduced Brillouin zone. This would be expected to produce pieces of the Fermi surface much smaller than those for the non-CDW state.

In the case of $4Hb$-TaS_2 no specific band-structure calculations exist; however, it is generally thought that the CDW transitions occur independently in the octahedral and trigonal prismatic layers[3,5] at 315 and 21 K, respectively. Because of this the conductivity at low temperature should be dominated by the trigonal prismatic layers separated by insulating octahedral layers. The Fermi surface at 1.1 K would result from the octahedral-layer superlattice with $\lambda_{CDW} = 13a_0$ and a transition in the trigonal prismatic layer similar to that occurring in the $2H$ polytype where a $3a_0$ superlattice results. A detailed model will require folding the band structure into the reduced zone in the presence of the triple CDW.

Rice and Scott[6] have carried out a preliminary band folding for a two-dimensional model of a triple-CDW state based on their saddle-point mechanism.[7] This results in a Brillouin zone reduced to $\frac{1}{9}$ of the original area, and the Fermi surface resulting from their band folding results in a number of small-area sections of Fermi surface consistent with the frequencies observed in the present experiment. Although such a comparison should be considered as only qualitative at this time, the many small frequencies observed in the present experiment are clearly consistent with a Fermi surface divided into sections by the gapping which results from the formation of a commensurate CDW superlattice. Preliminary de Haas–van Alphen measurements show oscillations at the same frequencies as observed in the transport measurements and these will be discussed in a later publication.

The novel behavior of the oscillatory voltage is a new effect and may be associated with the extreme anisotropy of the $4Hb$ phase where nearly insulating layers separate the metallic layers. For the various geometries this may introduce unusual boundary conditions with little or no current flow in the region of the potential contacts. However under these conditions large anomalous oscillatory voltage components are observed. The existence of tunneling[1] conductivity across the octahedral layers may also have to be considered. The effects are clearly coupled to the Landau levels and produce macroscopic voltages that indicate a coherent coupling over macroscopic lengths in the crystal. The magnitude of the oscillations are extremely large considering the measured residual-resistance ratios of ~200 for the best crystals. The two-dimensional character of the $4Hb$ phase could be playing a role in enhancing the oscillatory amplitude, or magnetic breakdown effects[8] can also contribute to amplitude enhancement. An interesting speculation concerns any possible role that the CDW state may play in either the large amplitude or the unusual current and field dependence of the oscillations.

This is the first observation of Shubnikov–de Haas effects in the magnetoconductivity of a layer-structure dichalcogenide although Graebner and Robbins[9] have been able to observe magnetothermal and de Haas–van Alphen oscillations in $2H$-$NbSe_2$. They have observed a frequency of 1.4 MG for $H_\perp$, and the angular dependence follows a $1/\cos\theta$ curve. They conclude that a pancake piece of Fermi surface is being observed with an average diameter of 0.344 $Å^{-1}$ and thickness 0.054 $Å^{-1}$. Further experimental observations of Fermi-surface effects in any of the phases of the layer-structure dichalcogenides would be most useful.

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