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ANOMALOUS MAGNETORESISTANCE IN CHARGE DENSITY WAVE COMPOUNDS: IS $NbSe_3$ UNIQUE?

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The partially gapped charge density wave (CDW) conductor $NbSe_3$ displays a dramatic magnetoresistance in the lower CDW state. We have made magnetoresistance measurements on the related partially gapped CDW compounds $ZrTe_3$ and $K_3Cu_8S_6$. Despite structural and electronic similarities of these materials to $NbSe_3$, we find non-existent or very small magnetoresistance effects. These results suggest that the anomalous magnetoresistance observed in $NbSe_3$ may be limited to partially gapped chain-like CDW conductors that contain a depinnable CDW.

I. Introduction

Highly anisotropic metallic compounds are susceptible to the formation of a periodic lattice distortion at moderate temperatures due to the strong electron-phonon interactions that stem from their low dimensional nature.[1] This distortion , known as a Peierls transition, gaps all or part of the Fermi surface. The carriers removed from the metallic states form a charge density wave (CDW) which does not take part in conventional ohmic transport.[2] Peierls transitions occur in both one-dimensional (1D) and two-dimensional (2D) metallic compounds. Due to their highly anisotropic nature, most chain-like quasi-one-dimensional compounds are completely gapped by the Peierls transition. They therefore become semiconductors in the CDW state. In many quasi-1D materials (such as TaS_3 and $(TaSe_4)_2I$) the CDW carriers can take part in collective, non-ohmic ("sliding") transport if an electric field strong enough (typically 10 to 1000 mV/cm) to counteract the effects of pinning impurities is applied along the CDW axis.[3]

Due to their reduced relative anisotropy, layered 2D compounds are only partially gapped by the Peierls transition. These materials stay metallic in the CDW state, albeit with a reduced number of carriers and hence a higher resistivity.[4] For most 2D CDW systems (such as TaS_2 and $NbSe_2$) the CDW is strongly pinned and cannot be made to slide by application of external electric fields. An interesting exception to this dimensionality/depinning trend is $NbSe_3$. Due to its complicated morphology, $NbSe_3$ is somewhere between 1D and 2D. The material undergoes two independent CDW transitions, neither of which entirely gaps the Fermi surface.[2] Despite its 2D character, the CDW in $NbSe_3$ is easily depinned (in fact, $NbSe_3$ has the lowest threshold electric field, ~1mV/cm, of any CDW conductor).

Recent studies[5] have demonstrated an anomalous magnetoresistance effect in the lower CDW state (T<59K) of $NbSe_3$. The application of a large magnetic field (~20T) directed perpendicularly to the chain axis is sufficient to more than double the low field resistance. The magnetoresistance is roughly linear in H-field, and is easily observed in moderate fields even below 3T. This unusual effect, which occurs at temperatures where conventional magnetoresistance mechanisms are inapplicable, has yet to be observed in any other CDW compound.

A model[6] which attempts to explain the anomalous magnetoresistance suggests that the magnetic field increases the 1D character of $NbSe_3$ so as to drive additional normal charge carriers into the CDW, resulting in a large increase in the normal carrier (ohmic) resistance. While various transport measurements provide indirect evidence for magnetic field induced carrier conversion,[7,8] more direct measurements of the CDW carrier concentration have thus far produced conflicting results.[9-11] Hence, the precise nature of the mechanism responsible for the anomalous magnetic effects in $NbSe_3$ is as yet unclear.

The low dimensional compounds $ZrTe_3$ and $K_3Cu_8S_6$ undergo Peierls transitions to the CDW state at moderate temperatures.[12,13] For both materials the transition only partially gaps the Fermi surface. Hence, as with $NbSe_3$, $ZrTe_3$ and $K_3Cu_8S_6$ are metallic down to 4K. Based on the magnetic field induced carrier converison model[6], these two compounds are potential candidates for showing anomalous magnetoresistance effects analogous to those observed in $NbSe_3$.

In this Communication we report the results of an experimental search for anomalous magnetic effects in $ZrTe_3$ and $K_3Cu_8S_6$. Magnetoresistance

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measurements performed on $ZrTe_3$ and $K_3Cu_8S_6$ in their respective low temperature CDW states indicate that neither material exhibits any dramatic magnetoresistance; only below roughly 15K does a substantial non-zero (isotropic) magnetoresistance become evident. We attribute this low-temperature effect to conventional magneto-transport mechanisms. We have also searched for possible electric field induced nonlinear electrical conductivity in the CDW states of $ZrTe_3$ and $K_3Cu_8S_6$. No evidence for a sliding CDW is found. These results, coupled with previous results on $NbSe_3$, define narrow constraints regarding the specific structural and electronic properties that are required for a CDW compound to exhibit anomalous magnetoresistance.

II. Experimental

Four probe dc conductivity measurements were performed on high quality single crystals of $ZrTe_3$ and $K_3Cu_8S_6$ prepared at the University of Kentucky and Cornell University, respectively. We refer the reader to Refs. 12 and 13 for a discussion of the synthesis techniques employed to grow the two compounds. Evaporated Indium contacts were used on $ZrTe_3$ crystals that had typical dimensions 2.0x0.6x0.1 mm^3. Gold conductive paint was employed to make contacts to $K_3Cu_8S_6$ samples that had typical dimensions of 0.1x0.3x0.1 mm^3.

Magnetoresistance measurements were performed in an 8T superconducting solenoid, with a small dc test current applied to the crystal. Large magnitude pulsed currents were appplied in zero magnetic field to test for non-ohmic I-V characteristics.

A. $ZrTe_3$

$ZrTe_3$ consists of metallic chains positioned along the b axis which are grouped in sheets in the a-b plane.[14] The interplane coupling is quite small ($\sigma_c \sim 0.1\ \sigma_{ab}$), while the interchain coupling is slightly larger that that along the chains ($\sigma_a \sim 1.2\ \sigma_b$).[12] $ZrTe_3$ forms in a Q2D layered structure and undergoes a Peierls transition at 63K; the transition removes only a portion of the Fermi surface. The resulting CDW is directed perpendicularly to the chain (b) axis, with a wave vector $q = 0.071\ a^*$.[15]

The a-axis resistance of $ZrTe_3$ below room temperature is shown in Fig. 1. The results are in qualitative agreement with previously published results.[12] The 63K Peierls transition is clearly evident in the data as an abrupt change in the slope of resistance versus temperature. The creation of the CDW results in the formation of a rounded anomaly in the resistance. $ZrTe_3$ continues to display metallic (R ~ T) behavior below 50K, albeit with an increased resistance. This indicates that the Peierls transition only partially gaps the Fermi surface.

Fig. 2 shows the detailed a-axis (CDW axis) resistance of $ZrTe_3$ measured with and without an applied H field of 7.5T. For the data shown, H was directed along the c-axis (and perpendicular to the a and b axes). Above about 20K, there is no observed magnetoresistance. Similar results are obtained for H applied along the a axis or along the b axis. In all cases, only below 20K does a small non-zero magnetoresistance appear, which grows monatonically with decreasing temperature. Hence, in $ZrTe_3$ only a conventional low temperature (isotropic) magnetoresistance is observed; for H-fields up to 7.5T we find no evidence for anomalous, anisotropic magnetoresistance at any temperature in the CDW state.

The I-V characteristics of $ZrTe_3$ were examined at selected temperatures in the CDW state below 63K. For electric fields directed

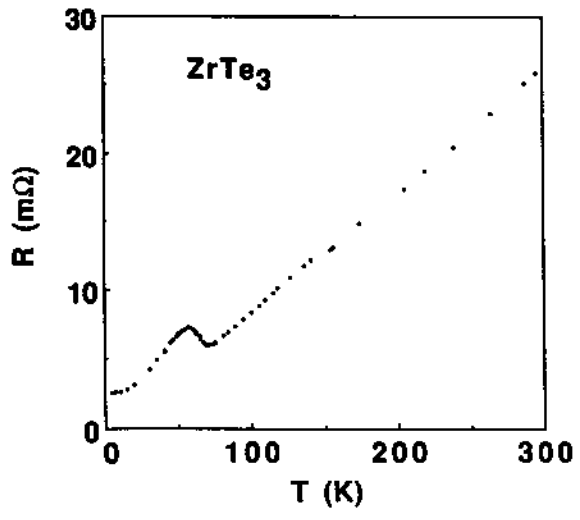

Fig. 1: Temperature-dependent resistance of $ZrTe_3$ along the a axis in zero magnetic field. The CDW transition is clearly evident as an upturn in R beginning at 63K.

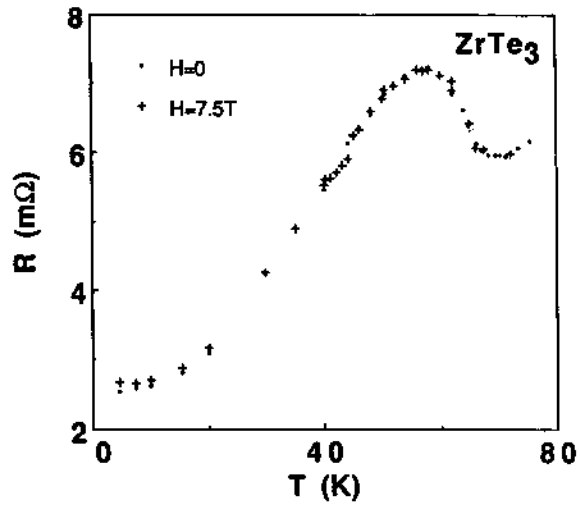

Fig. 2: The resistance of $ZrTe_3$ in the CDW state along the CDW (a) axis with H=0 (solid circles) and H=7.5T (crosses). The magnetic field was applied along the crystallographic c axis.

both parallel and perpendicular to the a-axis, no nonlinearities were observed up to 1V/cm, the maximum E-field applied.

B. $K_3Cu_8S_6$

$K_3Cu_8S_6$ is composed of Cu_4S_4 subunits which form chains along the b axis.[12] The chains form weakly coupled sheets in the a-b plane. Unlike $ZrTe_3$, the interchain coupling is far smaller than the intrachain coupling in $K_3Cu_8S_6$. Hence, $K_3Cu_8S_6$ is more of a Q1D chain compound than is $ZrTe_3$. $K_3Cu_8S_6$ undergoes a Peierls transition at 153K which again only partially gaps the Fermi surface and creates a CDW directed along the b axis. The CDW wave vector is $q = 0.46\ b^*$.[16] Recent X-ray measurements indicate that the CDW gradually approaches commensurability upon cooling below 150K, undergoing an incommensurate to commensurate (IC) transition at 50K.[16]

The zero magnetic field chain axis resistance of $K_3Cu_8S_6$ in the temperature range 20-300K is depicted in Fig. 3. The data are in general agreement with the previous results of ter Haar, *et al.*[13] except in the region of the IC transition where the hysteresis is far more pronounced in our samples. The Peierls transition is clearly evident in the data as an abrupt change in R vs T at 153K. dR/dT continuously decreases below 150K, as expected from a transition thatonly partially gaps $K_3Cu_8S_6$; complete gapping would produce activated behavior (i.e., dR/dT growing with decreasing temperature). $K_3Cu_8S_6$ remains metallic below 153K, but with a reduced number of charge carriers. A first-order IC transition is responsible for both the abrupt drop in R below 60K, and the hysteresis evident upon warming.[16]

The magnetic field dependent chain axis resistance for $K_3Cu_8S_6$ is shown in Fig. 4; the magnetic field was directed perpendicularly to the chain axis. We observe only an extremely small and isotropic magnetoresistance effect upon cooling from 80 to 50K. The effect is most pronounced ($\Delta R/R \sim 1\%$) for temperatures between 60K and 70K. No effect was observed upon warming from 75K to 80K. Hence, as with $ZrTe_3$, $K_3Cu_8S_6$ does not exhibit a dramatic high temperature magnetoresistance of the type exhibited by $NbSe_3$.

Electric fields of up to 10 V/cm directed along the CDW chain axis of $K_3Cu_8S_6$ were unable to depin the CDW throughout the temperature range 60-110K

III. Discussion and Conclusions

The absence of unconventional magnetic effects in the CDW states of either $ZrTe_3$ or $K_3Cu_8S_6$ has important implications for the unusual magnetic field effects displayed by $NbSe_3$. The overwhelming difference between $ZrTe_3$ and $NbSe_3$ is that $ZrTe_3$ is strictly a layered compound whereas $NbSe_3$ is quite chain-like. This suggests that anomalous magnetoresistance may be limited to Q1D CDW compounds.

$K_3Cu_8S_6$ is a Q1D compound yet it shows no magnetically induced effects. However, $NbSe_3$ contains a depinnable CDW, while $K_3Cu_8S_6$ does not. Additionally, the CDW in $K_3Cu_8S_6$ undergoes a gradual approach to commensurability as the temperature drops from 153K to 50K. This latter complication may very well be the cause of the

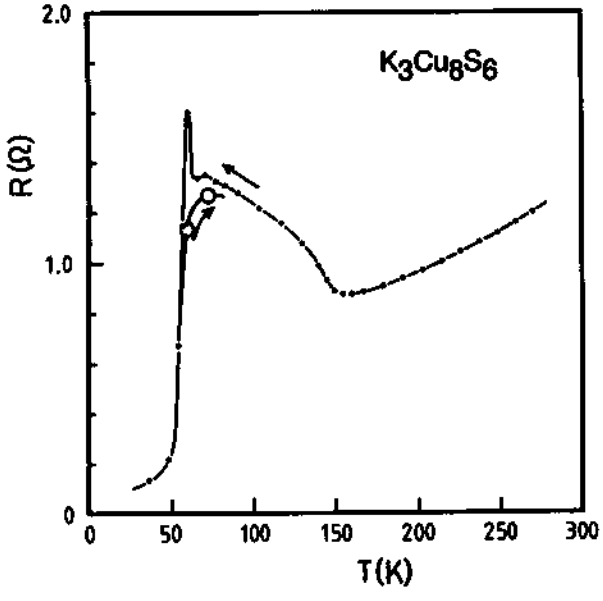

Fig. 3: Resistance vs. temperature for $K_3Cu_8S_6$ along the CDW (b) axis in zero magnetic field. The CDW transition at 153K manifests itself as an abrupt upturn in R. The arrows mark the warming (solid circles) and cooling (open circles) curves.

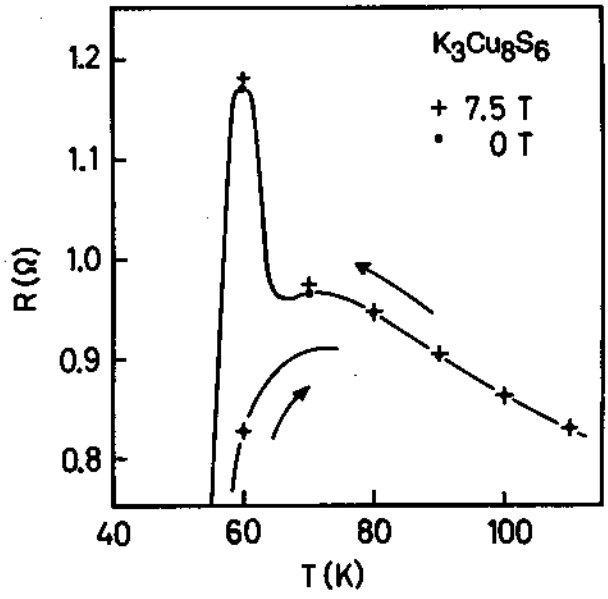

Fig. 4: Resistance vs. Temperature for $K_3Cu_8S_6$ along the CDW (b) axis for H=0 (closed circles) and H=7.5T (crosses). The arrows mark the warming and cooling curves. The magnetic field was applied perpendicularly to the CDW axis.

inability to depin the CDW in $K_3Cu_8S_6$. Hence, depinnability, which may be a manifestation of other attributes, is a possible second physical characteristic necessary for a material to exhibit anomalous magnetic effects.

We thus find that anomalous magnetic effects in CDW compounds may well be limited to those CDW materials which are sufficiently chain-like to allow collective transport yet not to so great an extent that the entire Fermi surface is gapped by the Peierls transition. This criterion defines an extremely narrow window in materials phase space. Presently, $NbSe_3$, with its unusual multichain structure, appears to be the only (inorganic or organic) material which falls within this restrictive range of parameters.

We next consider how these criteria relate to the model[5] of magnetic field induced carrier conversion. The limitations mentioned above suggest that a magnetic field is able to force carriers into the CDW, effectively enhancing the 1D nature of the compound, only if the material is very nearly an *inherently* one-dimensional compound. Hence, the CDW in a layered compound is presumably unaffected by an applied magnetic field because it is too two-dimensional. Similarly, chain-like materials which contain a non-depinnable CDW must contain sufficient 2D electronic structure to inhibit both CDW depinning and magnetic field induced carrier conversion. These results do not contradict the mathematical framework of the carrier conversion model as originally proposed by Balsiero and Falicov.[6]

In conclusion, we find no evidence for high-temperature, anisotropic magnetoresistance effects in the CDW compounds $ZrTe_3$ and $K_3Cu_8S_6$. These results strongly suggest that the anomalous magnetic field induced effects exhibited by $NbSe_3$ are limited to chain-like CDW compounds which show collective CDW transport and are only partially gapped by the underlying Peierls transition.

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