Angle-dependent magnetoresistance oscillations and Fermi surface reordering at high magnetic fields in \( \alpha-(ET)_2KHg(SCN)_4 \)

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Abstract

Angle dependent magnetoresistance oscillations (AMRO) have been studied in the charge transfer salt \( \alpha-(ET)_2KHg(SCN)_4 \) for magnetic fields in the range 0 - 30 T. This salt exhibits the onset of antiferromagnetic order at temperatures \( T_N \approx 8-10 \text{ K} \) and the presence below this temperature of a region of sharp negative magnetoresistance at a field around 22 T known as the “kink.” AMRO have been measured in this salt for a wide range of applied fields since the period, amplitude, and nature of the oscillations can be used to directly infer the character of the Fermi surface (FS) as a function of field. The data indicate that a profound change in the band structure occurs at this kink transition; the high field phase is characterised by quasi-2D oscillations from a closed cylindrical FS which is elongated in the \( c \) direction; the low field phase appears to be a spin density wave groundstate, with a FS consisting of a sheet (which is quasi-1D in character and tilted at an angle of \( \approx 21^\circ \) to the \( b^*c \) plane) and small closed 2D pockets. It is suggested that the breakdown orbits between the pockets and the 1D sheets are able to account for the various Shubnikov-de Haas frequencies observed below the kink.

1. Introduction

Charge-transfer salts of the form \( ET_2X \) (where \( ET \) is bis(ethylenedithio)tetrathiafulvalene and \( X \) is a monovalent anion) have been the subject of intense experimental study since high quality single crystals became available.\(^1,2\) The ET molecules are linked to each other by overlap of their molecular \( \pi \)-orbitals and stack along side one another, separated by sheets of the anion \( X \), to form a two-dimensional (2D) conductive network. Within this family of materials \( \alpha-ET_2MHg(SCN)_4 \) (\( M=K, Tl, Rb \) or \( NH_4 \) ) were synthesised as modifications of \( \kappa-ET_2Cu(SCN)_2 \) in an attempt to obtain a higher superconducting transition temperature \( (T_c) \).

The salts are isostructural (the so-called \( \alpha \)-phase) and as a consequence have almost identical predicted Fermi surfaces (FS) consisting of a 2D closed hole pocket and a pair of 1D planar FS sheets. The salt with \( M=NH_4 \) is a superconductor, with \( T_c \approx 1 \text{ K} \) The salts with \( M=K, Tl \) and \( Rb \) remain metallic down to \( < 100 \text{ mK} \) and all show the onset of antiferromagnetic order at temperatures \( T_N \approx 8-10 \text{ K} \) with the easy axis in the highly conducting \( ac \)-plane.

An interesting transition has been observed in the magnetic field dependence of the low temperature resistance of the salts with \( M=K \) and \( Tl \), but not with \( M=NH_4 \); the resistance increases with magnetic field up to \( \approx 10 \text{ T} \) (at 0.5 K), but then decreases with a region of very sharp negative magnetoresistance at \( \approx 23 \text{ T} \) (known as the ‘kink’). Above this field, Shubnikov de Haas oscillations are observed superimposed upon a monotonically increasing resistance (in contrast, the \( NH_4 \) salt exhibits this behaviour at all fields, not just above 23 T). In the \( K \) salt, the magnetoresistance exhibits significant hysteresis below the kink\(^4-7\), particularly when the \( ac \)-plane of the sample is tilted with respect to the magnetic field\(^2\).

It has been suggested that this kink transition is the point at which the external field destroys the low temperature antiferromagnetic state\(^5\), but the low temperature bandstructure and the field-induced changes giving rise to the kink have remained the subjects of speculation.

2. AMRO experiments

We have measured angle-dependent MR oscillations (AMRO) in single crystals of \( \alpha-(ET)_2KHg(SCN)_4 \) in magnetic fields up to 30 T in order to provide information about the FS topology below and above the kink. Magnetic fields up to 30 T were provided by the Nijmegen Hybrid II magnet. Standard 4-wire AC techniques (5-150 Hz) were used for all measurements, with the current directed in the inter-plane \( b^* \) direction.\(^2\)

The AMRO at 15 T (Fig. 1) indicate that the low magnetic field bandstructure is dominated by a 1D open
section of FS inclined at \( \sim 21^\circ \) to the crystallographic \( b^*c \) plane; qualitatively similar behaviour has also recently been observed in \( \alpha\text{-ET}_2\text{TIHg(SCN)}_4 \).\(^7\)

The action of the proposed SDW state\(^2\) on the calculated bandstructure results in a warped quasi-1D FS tilted by 26° with respect to the \( b^*c \) plane, plus small 2D pockets. Above the kink, the magnetoresistance is dominated by 2D orbits indicating a closed cylindrical FS. This transition is seen particularly clearly in Fig. 2 which shows AMRO at fixed \( \phi \): as the applied magnetic field is increased through the kink transition, dips in the resistance disappear and new peaks appear. This is because the two effects are different in origin: the minima at low field are due to a resonance/commensurability effect of electrons moving across the weakly corrugated 1D sheets\(^6\); the peaks in the high field AMRO result from the vanishing of the electronic group velocity perpendicular to the 2D layers and are connected with the corrugation of the quasi-2D cylinders.\(^9\)

Acknowledgments

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FIG. 1. AMRO of an \( \alpha\text{-ET}_2\text{KHg(SCN)}_4 \) crystal at 1.5 K at 15 T as a function of angle \( \theta \) and \( \phi \). The traces are offset for clarity.

On raising the magnetic field through the kink, however, the AMRO change in character (Fig. 2), indicating that the FS now possesses a 2D closed section in the form of a distorted cylinder.

FIG. 2. AMRO in \( \alpha\text{-ET}_2\text{KHg(SCN)}_4 \) at 1.5 K. The sharp dips seen at 17 T are seen at all lower fields, but change in character in the range 22–25 T.

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