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The low-temperature phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$: I. Angle and temperature dependence of the Shubnikov–de Haas and de Haas–van Alphen oscillations

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Abstract. The magnetoresistance and magnetization of single crystals of the organic charge-transfer salt α-(BEDT-TTF)$_2$KHg(SCN)$_4$ (where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene) have been studied in fields of up to 30 T and at temperatures as low as 20 mK. Five separate series of quantum oscillations have been observed in the low-temperature, low-field phase of this material and have been studied as a function of tilt angle of the field. It is proposed that two of these frequencies are the result of Stark quantum interference while the others are Shubnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) oscillations due to closed Fermi surface pockets or conventional magnetic breakdown. The unconventional temperature dependence observed for some of these oscillations and the applicability of current models of the Fermi surface of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ are discussed.

1. Introduction

The isostructural charge-transfer salts α-(BEDT-TTF)$_2$MHg(SCN)$_4$ (where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene and M = K, Rb, Tl or NH_4) have attracted considerable attention because they are quasi-two-dimensional (Q2D) metals and their low-temperature ground state can be one of two types, either superconducting (for M = NH_4) or characterized by what is thought to be a spin-density-wave (SDW) (for M = K, Tl or Rb) [1–7]. Measurements of the Fermi surface shape of these materials, using de Haas–van Alphen (dHvA) and Shubnikov–de Haas (SdH) oscillations, have been extremely useful in understanding the detailed differences between these salts [8–17]. Further information has been obtained by studying angle-dependent magnetoresistance oscillations (AMROs) which are very different for the two types of ground state [13, 18–21].

The picture which has emerged from these studies can be summarized as follows: in the superconducting salt (M = NH_4) the Fermi surface consists of a pair of warped quasi-one-dimensional (Q1D) sheets and a quasi-two-dimensional (Q2D) pocket; in the SDW salts (e.g. M = K) the Q1D sheets nest, leaving a reconstructed Fermi surface containing strongly corrugated sheets and some further pockets. The unreconstructed Fermi surface

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appears to be recovered in the SDW salts by warming above $T_N \sim 8$–10 K or applying a magnetic field higher than $B_K \sim 23$–35 T (known as the 'kink field' [13, 22]). The quantum oscillations in the M = NH_4 salt consist of a single frequency $F_\alpha = 567 \pm 1$ T [9] corresponding to the single Q2D pocket; in the M = K salt the quantum oscillations are extremely complicated, comprising several frequencies [11, 13, 14, 20], some of them reported to be sample dependent [11, 13], and with significant hysteresis between rising and falling field sweeps [11]. The precise origin of these frequencies has not been explained.

In this paper we report measurements of the angle dependence of the SdH oscillations in the M = K salt for temperatures in the range 0.5–4.2 K and fields of up to 30 T. These are compared with dHvA oscillations in this salt for temperatures in the range 0.02–2 K and in fields of up to 13.5 T measured by the field modulation method. This has allowed magnetic quantum oscillations in the reconstructed Fermi surface to be explored in much greater detail than has previously been possible. In contrast to earlier suggestions [11–13] we find that the quantum oscillation frequencies are in fact sample independent but that the amplitudes of the various series of oscillations do depend on sample preparation and/or cooling. This observation allows us to explain previous experiments [11–13] in a consistent manner. We also show that the SdH data exhibit additional series of quantum oscillations in comparison to the dHvA data. This has led us to propose that these extra oscillations can be attributed to the Stark quantum interference mechanism.

This paper is the first of a series of two, the second of which will report on measurements of the SdH effect in both α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ when hydrostatic pressure is applied [23]. Throughout these articles we shall refer to the present paper as I and the following one as II.

2. Experimental method

Single-crystal samples of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ were prepared using standard electrochemical techniques [2]. The resultant black platelets had 25 μm diameter gold wires attached to the faces that corresponded to the crystalline *ac* planes. Standard four-wire a.c. techniques (131 Hz) were then used to measure the sample resistance. A current of 5 μA was passed through the crystals in the direction of the (interplanar) b^* axis. The magnetoresistance measurements were performed in a ^{3}He cryostat that allowed the sample to be rotated about its *a* axis (i.e. with the magnetic field in the *bc* plane) with a precision of $\pm 1^\circ$. Fields of up to 30 T were provided by Bitter and superconducting magnets. Several crystals have been used in the magnetoresistance measurements; for consistency's sake, the magnetoresistance data shown in the figures have all been taken using the same crystal which has also previously been employed in the extensive study of the AMROs in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ reported in [19].

The dHvA oscillations of a crystal of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ were also studied using the field modulation method in a dilution refrigerator equipped with a 13.5 T superconducting magnet [24]. The angle dependence of the dHvA oscillations were studied at a temperature of 100 mK for angles of up to $\theta = 65^\circ$ where θ is the angle between the crystalline b^* axis and the field direction.

3. Experimental data

The SdH and dHvA waveforms of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ recorded with the field perpendicular to the *ac* plane ($\theta = 0^\circ$) are presented in figures 1(*a*) and 1(*b*) respectively. In both cases the quantum oscillations are complex but dominated by a frequency of (671±3) T (the α frequency) which shows a pronounced second-harmonic component. The SdH data in particular show a definite beat structure in the envelope of the oscillations; we shall initially consider the spectral content of the magnetoresistance data before turning to the dHvA data.

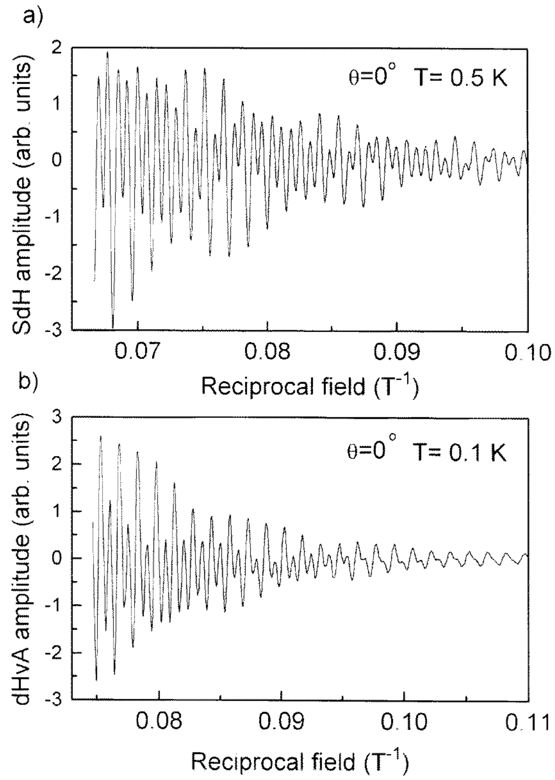

Figure 1. Oscillatory component of (*a*) the magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at 0.5 K and (*b*) the oscillatory component of the dHvA signal at 0.1 K with the magnetic field perpendicular to the ac plane.

3.1. SdH oscillations

As the sample was initially cooled the temperature was stabilized at a number of points at which magnetoresistance field sweeps were made. In figure 2 Fourier transforms show how the SdH oscillations evolve between 4.2 K and 0.5 K. Initially only two clear peaks are visible: one centred at 671 T (the α frequency) and one at 856 T (which we shall refer to throughout this paper as the ν frequency). By 2.3 K both of these peaks have increased in amplitude, although the amplitude of the α peak has increased at a faster rate than that of the ν peak. At this temperature the first hint of a third peak at $\sim$775 T (which will henceforth be referred to in this paper as the μ frequency) can be seen to emerge. Upon cooling to 1.5 K the α peak has grown further and becomes by far the most dominant SdH frequency. Meanwhile the μ peak has increased in size such that it is now clearly resolvable above the noise level but the ν frequency has *decreased* in amplitude.

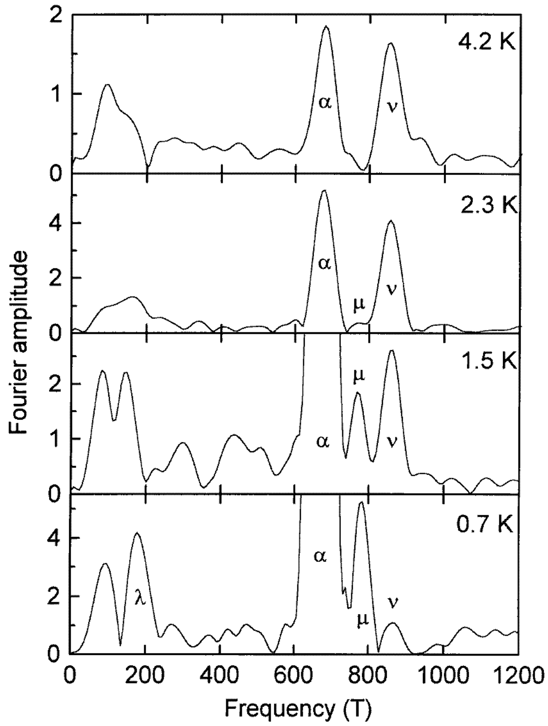

Figure 2. Temperature dependence of the Fourier transforms of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ magnetoresistance from 0.7–4.2 K.

The decrease in the amplitude of the ν frequency with cooling is contrary to the standard model of SdH oscillations embodied in the Lifshitz–Kosevich (LK) formula [25]. In this model the reduction in the thermal broadening of the Fermi–Dirac distribution as the temperature is lowered leads to more sharply defined oscillations in the density of states and hence an increased amplitude for the SdH oscillations. Such non-LK behaviour is not without precedent in BEDT-TTF salts; β''-(BEDT-TTF)$_2$AuBr$_2$ is also believed to possess a SDW ground state, and exhibits decreasing quantum oscillation amplitudes upon cooling [26]. The falling amplitude of the ν frequency thus requires further explanation and we shall return to this point later in this paper.

Finally, the Fourier transform of data at 0.7 K shows that the ν frequency has been suppressed to the extent that it is only just visible above the background noise. Both the α and μ peaks have continued to increase in size and a new peak at ∼181 T has emerged (we denote this the λ frequency). Although there are other small features in the Fourier transforms of figure 2, we will only be concerned with the λ (∼181 T), α (∼671 T), μ (∼775 T), ν (∼856 T) and β (see below) (∼4270 T) frequencies since these appear to be the only real quantum oscillatory features which we find repeatably in all samples that we have studied and which exhibit Q2D behaviour as the sample is tilted with respect to the field (see section 3.3).

In figure 3(*a*) a Fourier transform of the α-(BEDT-TTF)$_2$KHg(SCN)$_4$ SdH data shown in figure 1(*a*) is presented, displaying the higher-frequency region of the spectrum. Note that in this case it was necessary to plot the logarithm of the Fourier amplitude in order to accommodate all the spectral features on a single graph. Here the λ, α and μ frequencies are visible along with a new peak centred at ∼4257 T, which will henceforth be referred to as the β frequency. It is also noteworthy that the largest peak in the spectrum corresponds to the second harmonic of the α frequency while third and fourth harmonics of the α frequency are also clearly resolved. This aspect of the behaviour of the α frequency is not, however, duplicated by the λ, μ or (at higher temperatures) ν frequencies, which display no discernible higher harmonics.

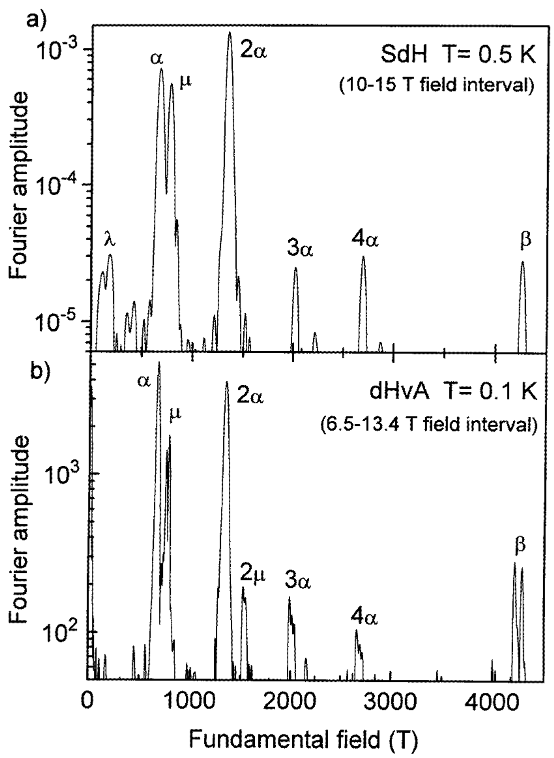

Figure 3. Fourier transform of (*a*) the magnetoresistance and (*b*) the dHvA signal of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ with the magnetic field perpendicular to the *ac* planes.

Previous workers have applied the LK formula to the temperature dependence of the SdH in the SDW phase of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in order to deduce the effective masses associated with the various frequencies [10, 13, 17]. There have been a variety of masses reported for the α series of oscillations, lying generally in the range 1.4–2.0m_e. Figure 4(*a*) shows a fit of the LK formula to the temperature dependence of the α-frequency SdH oscillations. The effective masses derived from the temperature dependence of the magnetoresistance oscillations are shown in table 1. The effective mass (m^*) estimated for the fundamental α frequency of the SdH data, in the temperature range 0.7–4.2 K, was $(2.0 \pm 0.1)m_e$. The effective mass associated with the second harmonic of this frequency was found to be $(3.2 \pm 0.5)m_e$, somewhat smaller than would be expected for conventional LK behaviour. A mass of $(1.8 \pm 0.1)m_e$ was also derived for the μ frequency from the SdH oscillations. Accurate estimates could not be made for the masses of the λ and β frequencies, or for other harmonics of the α frequency from the magnetoresistance measurements since these peaks were only resolvable near base temperature. An estimate of m^* for the ν peak

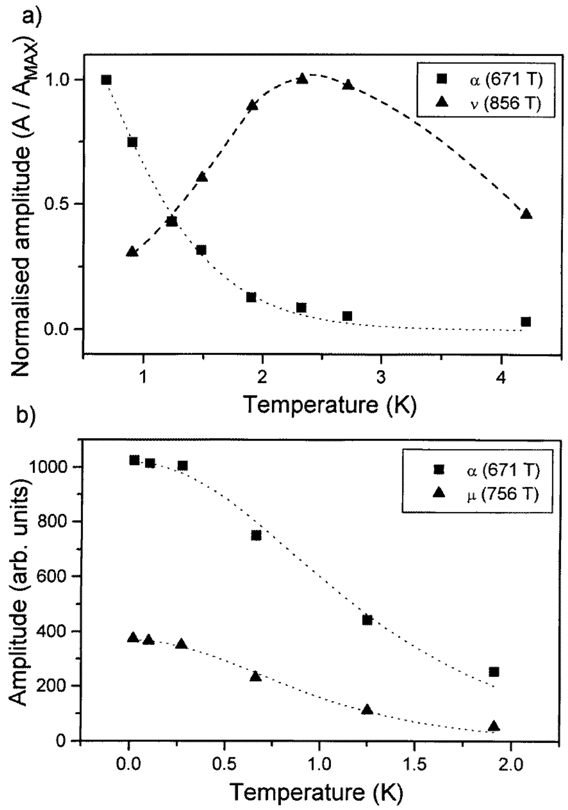

Figure 4. (*a*) Temperature dependence of the amplitudes of the α (squares) and ν (triangles) series of oscillations in the magnetoresistance. The dotted line is a fit of the LK formula to the α-frequency amplitudes while the dashed line through the ν-frequency points is merely a guide to the eye. (*b*) Temperature dependence of the amplitudes of the α and μ series of dHvA oscillations with fits of the LK formula (dotted curves).

was impossible due to its unusual temperature dependence (note that the dashed line through the ν-frequency data of figure 4(*a*) is merely a guide to the eye, not a fit).

3.2. dHvA oscillations

Figure 3(*b*) shows a Fourier transform of the dHvA oscillations in α-(BEDT-TTF)$_2$-KHg(SCN)$_4$ at 100 mK, with the field perpendicular to the *ac* plane ($\theta = 0°$). As in the SdH data, the α frequency dominates the Fourier spectrum, possessing a pronounced second harmonic and associated higher harmonics. The Fourier transform again contains components corresponding to the μ and β frequencies but in this case instead of a single Fourier peak at each frequency two peaks are resolved. The frequency μ is split into $\mu_1 = 756$ T and $\mu_2 = 783$ T while that at β becomes $\beta_1 = 4209$ T and $\beta_2 = 4283$ T. This splitting of the Fourier transform peaks is thought to be due to resolution at low magnetic fields of the extremal maximum and minimum orbits on the warped Q2D Fermi surface cylinders and/or breakdown orbits that give rise to these series of dHvA oscillations; this has been dealt with in detail in [24] and [27]. A similar splitting of the Fourier transform peak of the fundamental of the α-frequency SdH series is not resolvable but the peaks of

Table 1. Summary of the properties of quantum oscillations observed in the SDW state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$.

Series	F_0 (from magneto-resistance) (T)	F_0 (dHvA) (T)	Present in magneto-resistance	Present in dHvA	Present in low-field state	Present in high-field state	Effective mass, m^* in low-field state SdH	Effective mass, m^* in low-field state dHvA
λ	181 ± 3	—	Yes	No	Yes	No	—	—
α	671 ± 2	671 ± 3	Yes	Yes	Yes	Yes	2.0 ± 0.1	1.46 ± 0.1
2α	1342 ± 3	1340 ± 3	Yes	Yes	Yes	Yes	3.2 ± 0.5	2.9 ± 0.2
μ	775 ± 3	756 ± 5 (μ_1) 783 ± 5 (μ_2)	Yes	Yes	Yes	No	1.8 ± 0.1	1.7 ± 0.1
ν	856 ± 5	—	Yes	No	Yes	No	—	—
β	4270 ± 5	4218 ± 8 (β_1) 4284 ± 10 (β_2)	Yes	Yes	Yes	No	—	3.3 ± 0.5

its higher harmonics are split. The sixth harmonic, for example, is split into $6\alpha_1 = 3975$ T and $6\alpha_2 = 4055$ T. This corresponds to a splitting of $\sim$13 T for the fundamental.

In contrast, however, to the SdH data, no frequency equivalent to the λ frequency is observable in the dHvA oscillations down to temperatures as low as 20 mK. Furthermore, field sweeps at temperatures in the range 0.02–4.2 K revealed no dHvA frequency corresponding to the ν frequency.

Effective masses have been estimated for the dHvA frequencies observed in these data (figure 4(*b*)). The α frequency has $m^* = (1.46 \pm 0.1)m_e$ with its second harmonic having a mass of $(2.9\pm0.2)m_e$. The μ frequency has a mass of $(1.7\pm0.1)m_e$ and its second harmonic has a mass of $(3.2 \pm 0.4)m_e$. The mass estimated for the β frequency was $(3.3 \pm 0.5)m_e$.

It is noted that the effective mass of the μ frequency estimated from the SdH oscillations and that derived from the dHvA oscillations agree with each other within the experimental error. However, this is not the case for the effective masses estimated for the α-frequency series. Although the LK fits presented in figures 4(*a*) and 4(*b*) for this series of oscillations extend over different temperature ranges for the SdH (0.7–4.2 K) and dHvA (0.02–1.9 K) data sets we have also fitted the LK formula to the SdH and dHvA oscillations over the same temperature range (0.7–1.9 K). The latter procedure did not produce significantly different values from the fits carried out over the full temperature ranges. In previous experiments the effective mass estimates derived from SdH data have varied over the range 1.4–2.0m_e [10, 11, 13] and it has been speculated [13] that this inconsistency might be due to incorrect removal of the background magnetoresistance. In figure 4(*a*) the background magnetoresistance was removed by division of a polynomial fit of appropriate order. However, the results of other procedures, such as background subtraction, were found to yield similar effective mass values. The difference between the mass estimated from the LK analysis of the dHvA and SdH oscillations thus appears robust.

Recently it has been shown that it is inappropriate to apply the LK formula to the quantum oscillations occurring at high magnetic fields when there are coexisting sections of Q1D and Q2D Fermi surface, as is the case for this material [27]. Oscillations in the chemical potential, arising from its being pinned to very sharp Landau levels, bring about significant departures in the form of the temperature dependence of the oscillations from the predictions of the LK formula, in which the chemical potential is assumed to be constant. This manifests itself most obviously when the LK formula is used to analyse the temperature dependence of the higher harmonics of the dHvA oscillations; the effective mass m_p^* of the

pth harmonic thus deduced is smaller than the LK prediction pm_1^* [14, 27]. Furthermore, LK analysis of the SdH oscillations under these conditions may yield an artificially large apparent effective mass [27]. Detailed numerical simulations [27] have shown that under the high-field conditions masses extracted from the dHvA oscillations are a more reliable guide to the true effective mass than those derived from the SdH oscillations. Therefore this may indicate that when considering the discrepancy between the SdH and dHvA masses measured in the SDW state the dHvA value of $m_1^* \approx 1.4m_e$ is possibly the more reliable. We note that the effective masses deduced using LK analysis from the second harmonic of the α frequency in both SdH and dHvA oscillations are very similar. It is possible that in the SDW state the amplitude of the SdH oscillations may be further affected by phenomena such as spin-dependent scattering and therefore our effective mass estimation, via the LK formula, may serve only as a parametrization of the differences in scattering processes.

3.3. Angular dependence of the quantum oscillations

Figure 5 shows the quantum oscillation fundamental fields versus the tilt angle, θ, of the normal to the ac planes (b^*) relative to the applied magnetic field. In a Q2D system the frequency $F(\theta)$ of quantum oscillations as a function of tilt angle should be described by a curve of the form

$$F(\theta) = \frac{F(0)}{\cos\theta}. \tag{1}$$

This equation has been fitted to the data for the λ and μ frequencies derived from the 500 mK SdH field sweeps in figures 4(a) and 4(b) respectively. It is clear from these fits that the frequencies have the expected behaviour for features resulting from a Q2D Fermi surface. As can be seen in figure 3(a) the λ frequency is close to the noise limit of the Fourier transform at $\theta = 0°$. However, figure 6(b) contains a Fourier transform of a field sweep made at an angle of $\theta = 35°$ where a peak corresponding to λ is clearly resolved at ∼210 T with its second harmonic also present at ∼420 T. Generally the λ frequency was found to be best resolved at high tilt angles ($\theta \approx 30°$–$50°$). Similar behaviour of low-frequency SdH oscillations in α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ has been reported to occur at a tilt angle of $\theta \sim 45°$ [28]. Fits of equation (1) to the data of figures 4(a) and 4(b) yielded $F_\lambda(0) = (181 \pm 3)$ T and $F_\mu(0) = (777 \pm 3)$ T. A similar fit to the μ frequency found in the 100 mK dHvA data gave $F_\mu(0) = 775$ T.

Figure 5(c) shows a fit of equation (1) to the ν frequency at 1.4 K. Most of the angle-dependent field sweeps were carried out at low temperatures ($T < 500$ mK) where the ν frequency is not clearly resolvable in the Fourier transform, hence the limited number of data points for this plot. It is apparent, however, that the data are well modelled by a fit with $F_\nu = 856$ T.

The angle dependence of the β frequency is shown in figure 5(d). In this case the frequency was resolved into two separate peaks in the Fourier transforms of the 100 mK dHvA data. Fits have been made to the angle dependence of the lower of the two frequencies, β_1 (square data points) and the higher, β_2 (triangular symbols) to yield $F(0)_{\beta 1} = (4218 \pm 8)$ T and $F(0)_{\beta 2} = (4284 \pm 10)$ T.

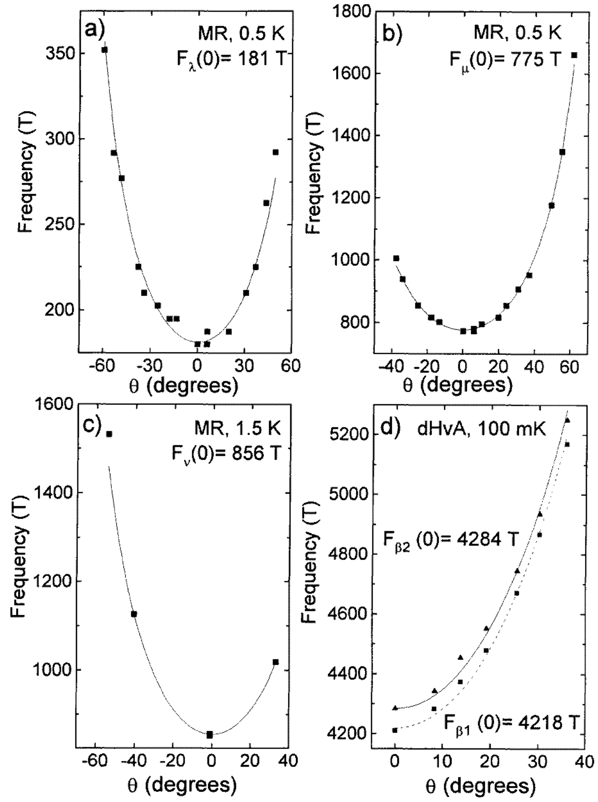

Figure 5. Angle dependence of the (*a*) λ frequency, (*b*) μ frequency, (*c*) ν frequency and (*d*) split peak β frequency in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. (*a*), (*b*) and (*c*) are derived from magnetoresistance measurements while (*d*) is from dHvA data at 100 mK.

4. Discussion

4.1. The λ and ν frequencies: Stark quantum interference

It has been established that there are multiple series of SdH oscillations present in the magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ within the low-field state. The properties associated with each series are summarized in table 1.

Firstly, it is noted that of the five separate frequencies of quantum oscillations observed in the magnetoresistance only the α, μ and β frequencies are seen in the magnetization (dHvA) data, the λ and ν frequencies being measurable only in the transport experiments. This suggests that the λ and ν frequencies are most likely not arising from carrier orbits on closed Q2D Fermi surface pockets or conventional breakdown orbits, as would be the case for frequencies seen in both dHvA and SdH oscillations. They may instead be due to a mechanism such as Stark quantum interference [29]. This occurs when two sections of Fermi surface lie close to each other, separated only by a narrow energy barrier, such that they almost form a closed Fermi surface loop. If the separation of the Fermi surface sections is small enough then carriers will be able to tunnel between them. Thus they will have a significant probability of traversing the loop by two alternative paths. In this event the carrier wavefunction will interfere with itself around the loop, this interference being dependent upon the phase shift caused by the magnetic flux enclosed within the corresponding real space loop. This leads to oscillations in the resistance of the sample that are periodic in a reciprocal field. Since the carriers never complete an orbit of a Fermi surface pocket the effect will not show up in the magnetization but may contribute to the magnetoresistance.

In the low-field, low-temperature state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ where the Fermi surface has been nested, it is likely that there will exist sections of Fermi surface in close proximity to each other [12]. Such pieces of Fermi surface may be the source of Stark quantum interference oscillations. This proposal is supported by the fact that neither the λ or ν frequency are observed in the magnetoresistance above the kink transition where the Fermi surface is thought to be in its unnested form.

Stark quantum interference oscillations can also be distinguished from the SdH effect because they are generally much less sensitive to temperature [29]. This means that, although such oscillations will also gradually reduce in amplitude as the temperature is raised, they will be expected to dominate Fourier transforms of the SdH data at the higher temperatures.

The ν frequency has been observed over the temperature range 0.7–4.2 K and found to have the unconventional temperature dependence described in section 3.1. At the highest temperatures that we have measured (4.2 K), the ν-frequency peak in the Fourier spectrum is comparable in magnitude to the α frequency and for some traces is actually the dominant peak. This supports the assumption that it may be a Stark oscillation. However, the reduction in the amplitude of the ν frequency observed at temperatures below $\sim$2 K is as unexpected for Stark oscillations as it would be for SdH oscillations. Since it is only the ν frequency that shows this reduction in amplitude at lower temperatures it seems likely that its suppression is due to a change in the Fermi surface restricted to the locality from which the ν frequency originates. This might, for example, be a very small change in the nesting of the Fermi surface such that the separation of the two sections responsible for the ν frequency increases enough to reduce the probability of tunnelling from one to the other to a negligible degree. The Q1D AMROs measured in this region of the phase diagram show no significant temperature dependence in the positions of their minima [13, 19], so any change in the topology of the Fermi surface sheets must be quite minor.

A minor alteration to the alignment of the Q1D Fermi surface sheets at temperatures below $\sim$2 K, might also explain why the λ frequency is only observed below $\sim$0.7 K, even though it appears to be a product of Stark quantum interference. Possibly the same change that suppresses the ν series of oscillations brings about the onset of the λ series. In connection with this point we note that the peaks occurring in the Fourier transform are related by the sum $\alpha + \lambda \approx \nu$ (although it must be remembered that in spite of this apparent relationship the λ and ν frequencies are not simultaneously present in the Fourier transform at any temperature). Such an additive relationship between the frequencies suggests that they may be linked topologically on the Fermi surface by a magnetic breakdown network which undergoes a small change near 2 K, such that the λ path becomes favoured over that of the ν frequency.

A further noteworthy aspect of the λ and ν frequencies is to be found in their angle dependence. As noted earlier and shown in figure 6(*b*), the λ Fourier transform peak at tilt angles in the region $\theta \approx 30^\circ$–50° is much more pronounced than that seen at other angles and becomes comparable in amplitude to the α-frequency peak. The amplitude of the Stark oscillations in magnesium have been shown to be sensitively dependent on the exact orientation of the crystal in the magnetic field [29]. It seems likely that this characteristic of Stark orbits may be responsible for the enhancement of the amplitude of the λ oscillations at high tilt angle. The magnitude of the Stark oscillations in α-(BEDT-TTF)$_2$KHg(SCN)$_4$

is not so strongly angle dependent as that observed in magnesium. This is to be expected for oscillations that originate from Q1D and/or Q2D Fermi surface sections rather than a more three-dimensional Fermi surface topology.

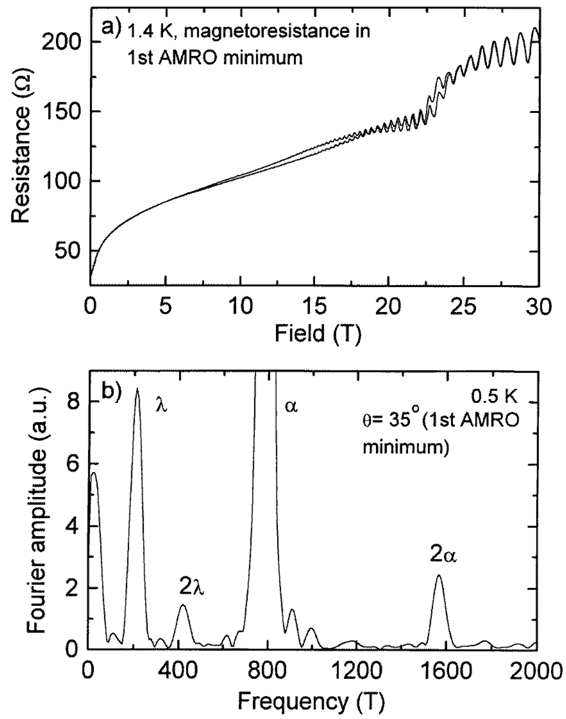

Figure 6. (*a*) Magnetoresistance (rising and falling field sweeps) of α-(BEDT-TTF)$_2$KHg (SCN)$_4$ at an angle corresponding to the first minimum in the SDW state AMROs. (*b*) The large λ-frequency peak seen in the Fourier transform of the magnetoresistance (field range 10–15 T) at this angle.

Large low-frequency ($\sim$15 T) oscillations have been observed to appear at a tilt angle of $\theta \approx 45^\circ$ in the magnetoresistance of α-(BEDT-TTF)$_2$TlHg(SCN)$_4$ by Brooks *et al* [28]. In view of the similarities between that material and α-(BEDT-TTF)$_2$KHg(SCN)$_4$ it seems likely that Stark quantum interference effects may also be responsible for those oscillations.

4.2. The μ frequency

The μ frequency differs from the λ and ν frequencies in that it is detectable in the magnetization measurements. It shares with these other frequencies the common property that it is only observed in the nested state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ i.e. it is not observed above the kink field [14]. For this reason it might be supposed that it is due to imperfect nesting of the unreconstructed Fermi surface, a mechanism that has been proposed to be responsible for the formation of certain Fermi surface pockets in β''-(BEDT- TTF)$_2$AuBr$_2$, another salt in the BEDT-TTF family that is believed to possess a SDW ground state [30].

Since this frequency is proposed to have its origins in a closed Q2D section of Fermi surface it might be expected to behave in a similar way to the α series of oscillations which are themselves conventionally associated with the Q2D section of the calculated Fermi surface. However, there are a number of differences in the behaviour of these two frequencies.

Firstly the μ frequency has much lower harmonic content than the α frequency. In our SdH measurements the μ series of oscillations had no clear harmonics while in the dHvA experiments only the fundamental and second harmonic were observed in Fourier transforms. In contrast, the α frequency had harmonics up to the fourth clearly present on Fourier transforms of the SdH data while in the magnetization data the sixth harmonic was observed. This difference between the two series is not just a result of the larger magnitude of the α oscillations but is a reflection of the extra harmonic content in the α series brought about by the large, very visible second-harmonic component that occurs in the magnetoresistance below the kink transition. One explanation of the strong second-harmonic content of the SdH oscillations involves exchange enhancement of the spin-splitting by interaction of the conduction electrons with the SDW moment [15]. In this model it is not clear why this mechanism should affect the α frequency but not the μ series. This point will be discussed in greater detail in paper II.

A second difference is seen in the estimates of the effective masses for these oscillations. Masses of $\sim$1.7–1.8m_e are obtained from both the SdH and dHvA measurements for the μ frequency (see table 1) whereas in the case of the α frequency the mass is $\sim$1.5m_e in dHvA and $\sim$2.0m_e in the SdH measurement. The reasons for this are unclear but we note that the presence of the spin-splitting of the α oscillations (but not the μ) below the kink transition and a simultaneous increase in Dingle temperature of the α frequency upon entering the SDW state [14] suggest that the α series may be affected by some type of magnetic scattering mechanism that is associated only with that particular section of Fermi surface.

4.3. Sample dependence of the λ, μ and ν frequencies

Previous workers have suggested that there are sample-dependent frequencies present in the quantum oscillations occurring in the magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. Pratt *et al* [11] reported a single sample-dependent frequency that occurred at $\sim$790 T in one sample and $\sim$860 T in another. In the light of our temperature-dependent SdH data on a single sample of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ it seems probable that Pratt *et al* were in fact measuring the μ frequency in one case and the ν frequency in the other, with differences in temperature or cooling procedure making one more pronounced than the other under slightly different conditions. Having now measured many different samples we have established that the position of the μ and ν peaks in the Fourier transform is sample independent but does vary with temperature as noted earlier. Pratt *et al* [11] also observed frequencies in the range $\sim$50–200 T. We suggest that these frequencies are a mixture of observation of the λ series at low temperatures and spurious peaks on the Fourier transform arising from the background magnetoresistance.

Although the peak positions in the Fourier transform have been established to be sample independent, we do not preclude the possibility that the relative magnitudes of the peaks may depend on cooling conditions. Observation of hysteresis in the magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ has led to the suggestion that the SDW state is composed of a magnetic domain structure consisting of regions that have undergone Fermi surface reconstruction and regions that remain unnested [16]. This model of the behaviour of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is deficient in a number of respects that will be discussed later in this paper. However, a model of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ including some form of domain structure (say perhaps with all of the separate domains being in SDW states but with small differences in their exact nesting vectors) provides a credible means of accounting not only for the magnetoresistance hysteresis but also variations in the relative content of the λ, μ and ν frequencies in the data. In this circumstance the exact cooling procedure might foreseeably alter the relative contributions of the different frequencies to the Fourier transform. Measurements by other groups have led to results where the ν and λ frequencies but not the μ have been observed [12, 13] and also results where only the α and β frequencies occur [17]. We propose that non-uniformity of cooling procedures and/or sample preparation between different groups may be responsible for these discrepancies.

4.4. The β frequency: magnetic breakdown

The β frequency occurs in both the SdH and dHvA quantum oscillations and is clearly Q2D as shown by its angle dependence (figure 5(*d*)). It is remarkable in that it has a fundamental field of ∼4270 T (see table 1), a frequency indicating a $\boldsymbol{k}$-space area almost exactly the size of the unreconstructed first Brillouin zone of α-(BEDT-TTF)$_2$KHg(SCN)$_4$.

Uji *et al* [17] have explained this frequency as corresponding to a large magnetic breakdown orbit that encompasses the unreconstructed Fermi surface. The observation of the β frequency, both in the present work and in results obtained by previous workers, has been made well within the phase boundary of the SDW state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ where the Fermi surface is believed to be nested. The size of the first Brillouin zone of the proposed nested α-(BEDT-TTF)$_2$KHg(SCN)$_4$ Fermi surface seems to be much too small to accommodate an orbit the size of the β frequency [12] and so models that have attempted to explain the presence of the β frequency have generally relied upon the assumption that there exists a domain structure whereby regions of the sample retain an unreconstructed Fermi surface even below the kink field. If such an assumption were correct then it would be expected that the β frequency would be most pronounced above the kink field, where all of the sample would be in the unnested state and the higher fields would lead to the magnetic breakdown orbit becoming progressively more dominant in the data. In this case, one might also expect to observe a β frequency in the quantum oscillations of the isostructural salt α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ which possesses no SDW state. This, however, is not the case. A recent dHvA study of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in fields of up to 54 T and temperatures down to 350 mK detected no β frequency above the kink transition [14]. It is notable that in the same measurement high-frequency (∼60 kT) dHvA oscillations from the polycrystalline copper in the pick-up coils were detected, indicating the very high sensitivity of this experiment to high-frequency dHvA oscillations. In a recent pulsed field study of the magnetoresistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$, Brooks *et al* [31] reported the observation of a β frequency in the SdH data at ∼4212 T. In the Fourier transform that they present the frequency peak corresponding to these oscillations is of comparable amplitude to the noise level. As such, we do not believe that this peak can be reliably ascribed to the β frequency. The only clear observations of this frequency have all been made in the SDW state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and therefore we suggest that the β frequency is a feature of this state alone (see also [32] and further discussion in paper II).

4.5. Models of the Fermi surface of α-(BEDT-TTF)$_2$KHg(SCN)$_4$

As has been discussed in the preceding section, the results presented in this paper pose interpretational difficulties for the current models of the Fermi surface of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. The proposal by Kartsovnik *et al* [12] of a SDW ground state with a nested Fermi surface existing below the kink field is still the model that best explains the general qualitative features of the transport properties of α-(BEDT-TTF)$_2$KHg(SCN)$_4$. In particular, this model is helpful in understanding the transition in the dimensionality of the AMROs at the SDW phase boundary [19], although it is insufficient to explain the detailed nature of the quantum oscillations that occur below the kink field.

If, as we have asserted in the present work, many of the quantum oscillations are either Stark quantum interference oscillations or involve imperfect nesting of the original Q1D Fermi surface sheets, then the precise topology of the Fermi surface in the proposed SDW state may be difficult to predict due to its sensitive dependence on the nesting vector and the exact shape of the Fermi surface in the unnested state. For this reason detailed AMRO experiments outside the SDW phase may be vital in deducing the reconstructed Fermi surface geometry [19].

An extension to the proposal of Kartsovnik *et al* [12] was suggested by Athas *et al* [16] who proposed a domain model whereby within the SDW state of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ there coexisted regions of the sample characterized by the unreconstructed Fermi surface, as well as others possessing nested Fermi surfaces. It was suggested that the quantum oscillations occurring in the nested state were a result of magnetic breakdown and that the breakdown probability became vanishingly small at special tilt angles of the magnetic field. These special angles corresponded to the minima in the AMROs found in the SDW state. The magnetoresistance within these minima was measured up to 20 T and found to have much in common with the magnetoresistance of α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$, in particular an absence of a spin-splitting waveform in the α frequency and a sublinear background magnetoresistance rather than the large hump seen in the magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at AMRO maxima.

During the experiments detailed in this paper we carried out AMRO sweeps so that we could align our sample exactly in the minima of the AMROs of the SDW state. At these points field sweeps were performed up to 30 T, as shown in figure 6(*a*); figure 6(*b*) contains a Fourier transform of SdH oscillations in an AMRO minimum, at 0.5 K. It is clear that this trace is similar to that obtained by Athas *et al* [16] up to 20 T. However, at $\sim$23 T a pronounced jump in the magnetoresistance is observed corresponding to the kink transition. This observation of the kink transition in an AMRO minimum is contrary to the idea of Athas *et al* [16] who propose that the properties of the reconstructed Fermi surface should not manifest themselves at this angle. It should also be noted that there is large hysteresis in the magnetoresistance between the rising and falling field traces, another illustration that magnetic ordering is still relevant to the transport properties of the sample within the AMRO minimum.

Further to this, Fourier transforms of data taken at this and other AMRO minima reveal the presence of the λ and ν frequencies (figure 6(*b*)). As mentioned earlier we believe that these frequencies are products of the reconstructed Fermi surface. It is thus clear that, contrary to the predictions of the model of Athas *et al* [16], many characteristics of the magnetoresistance that we identify with the SDW state persist within the AMRO minima.

5. Conclusion

The SdH and dHvA effects have been studied in single crystals of α-(BEDT-TTF)$_2$-KHg(SCN)$_4$ for a range of tilt angles of the sample relative to the applied field. Five different series of quantum oscillations have been observed within the low-temperature, low-field phase of this material where it is proposed that the Fermi surface derived from room-temperature crystallographic data is reconstructed by the formation of a SDW state.

The characteristics of these series of quantum oscillations are summarized in table 1.

Two of the frequencies (λ and ν) are not observed in the dHvA experiments and are ascribed to the Stark quantum interference mechanism occurring between Fermi surface sections that exist in the SDW state. An unconventional temperature dependence of these oscillations has been established and is proposed to result from minor changes to the nesting properties of the SDW at $\sim$2 K.

The α, μ and β frequencies are measurable in magnetization experiments and are thus assigned to closed Fermi surface pockets and/or breakdown orbits. These orbits are associated with the reconstructed α-(BEDT-TTF)$_2$KHg(SCN)$_4$ Fermi surface. Explanations for the existence of these frequencies are discussed in the context of current models of the Fermi surfaces of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ and it is noted that some of the models that have been proposed must be disregarded on the basis of incompatibility with the experimental data.

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