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Electron–electron correlations in $(BEDT–TTF)_2I_3$ organic superconductors

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The detection of the fractional Landau level filling factor $\nu = 1/2$ and low integer filling factors in the two-dimensional multilayer organic metal κ-$(BEDT–TTF)_2I_3$ is presented, which shows the occurrence of electron localisation and electron–electron correlation in this bulk metallic two-dimensional system. These effects are found in the normal conducting state of the organic superconductor κ-$(BEDT–TTF)_2I_3$. In addition, quantum oscillation measurements are found to be a very promising tool for direct detection of the chemical potential and its variation with magnetic field, even under rather complex fermiological conditions.

Key words: *BEDT–TTF; organic metals; Shubnikov–de Haas; de Haas–van Alphen; effective mass; Fermi surface*

1. Introduction

Radical salts of the electron donor BEDT–TTF (i.e. bis(ethylenedithio)-tetrathiafulvalene) with I_3^- anions are of a special interest, since their syntheses reveal a variety of electronic systems with identical stoichiometry, i.e. $(BEDT–TTF)_2I_3$, but with different structures (the so-called α-, β-, κ-, θ-, … phases). The α-phase is an organic–metal between room temperature (RT) and 135 K, with a sharp metal –insulator transition at this temperature [1], whereas the β-phase has metallic properties between RT and about 1 K, at which it becomes superconducting [2]. The κ- and θ-phases are both organic metals down to about 4 K, at which they become superconducting as well [3, 4]. Within the I_3 salts, the highest superconducting transitions at

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T_c = 8 K and ambient pressure were observed in α_t-(BEDT–TTF)$_2$I$_3$ [5] and β_H-(BEDT–TTF)$_2$I$_3$ [6], which are obtained from the initial α- and β-phases, respectively, by a special treatment of these materials.

In principle, some electron–electron correlation exist in all these different structural phases of (BEDT–TTF)$_2$I$_3$, as can be seen, for example, from strongly enhanced susceptibilities or effective masses of the carriers. In this paper, we will concentrate on electron–electron correlation in κ- and *θ*-(BEDT–TTF)$_2$I$_3$. Both these phases are not easily obtained in typical electrochemical process [1], but their syntheses have been brought further since they are of a special interest, e.g. with respect to their electronic dimensionality: the former κ-phase is probably the most extreme two-dimensional (2D) organic metal [7, 8] in the class of BEDT–TTF radical salts (the ratio of transfer integrals perpendicular and parallel to the conducting (*b*,*c*) planes is $t_\perp/t_{||} < 1{,}5\cdot10^{-4}$). The latter *θ*-phase is electronically quasi-two-dimensional (Q2D) and is the only BEDT–TTF radical salt that has a very small 3-dimensional (3D) closed orbit on the Fermi surface [9]. Moreover, *θ*-(BEDT–TTF)$_2$I$_3$ shows a strong magnetic interaction at high magnetic fields [10].

2. Experimental

κ- and *θ*-(BEDT–TTF)$_2$I$_3$ single crystals were synthesised by the usual electrochemical procedure [1]. Quantum oscillation (QO) experiments were performed as both de Haas-van Alphen (dHvA) and Shubnikov-de Haas (SdH) measurements in superconducting magnets (up to 10 T), as well as in resistive magnets up to 28 T provided by the Grenoble High Magnetic Field Laboratory. Different sample-contacting methods were applied in order to avoid subtle contact effects. Annealed gold wires with a thickness of 15–25 μm where applied either directly to the samples or on evaporated gold contacts. The wires were attached by gold, platinum or carbon paints, respectively. Low metallic contact resistances of about 2–5 Ω (at room temperature) could be obtained, even if the evaporation of gold was refused. ac currents of frequencies between 90 Hz and 4 kHz were applied perpendicular and parallel to the conducting planes and were limited to 50–300 μA. Low temperatures down to 0.38 K were realised by pumping on both a ^{4}He bath cryostat and its ^{3}He insert, whereas temperatures down to 20 mK were realised in a ^{3}He/^{4}He dilution refrigerator. Angle-dependent QO experiments were carried out by mounting the samples on a rotatable sample-holder.

3. Results and discussion

3.1. κ-(BEDT–TTF)$_2$I$_3$

Highly conducting (*b*,*c*) planes of κ-(BEDT–TTF)$_2$I$_3$ are denoted as 2D planes in the following. It should be emphasised that a typical bulk single crystal contains about

10^5 successive conducting layers. This is a typical order of magnitude within these so-called organic charge-transfer (CT) salts. The investigations presented here concentrate on the normal conducting state of this compound.

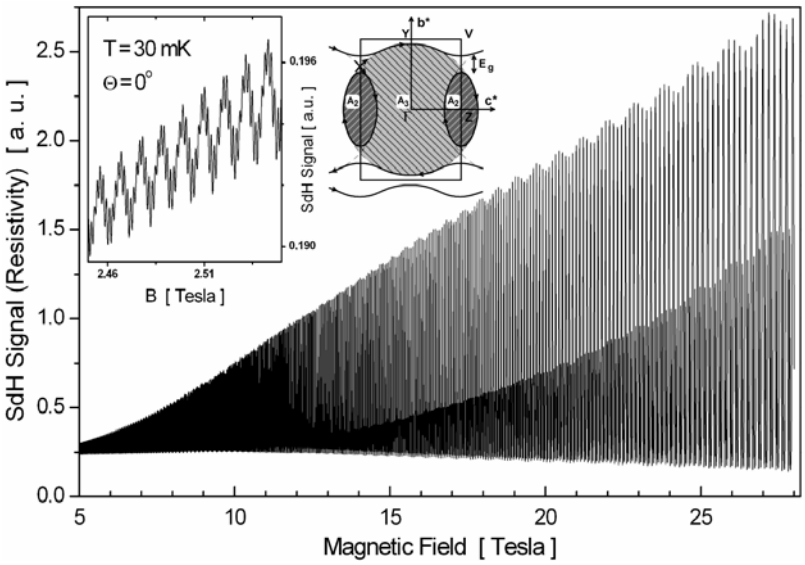

Fig. 1. Magneto-resistance and Fermi surface of κ-(BEDT–TTF)$_2$I$_3$. The inset shows the low-field part. For data evaluation, the SdH signal has to be divided by the non-oscillatory background magneto-resistance

Figure 1 shows typical SdH oscillations of κ-(BEDT–TTF)$_2$I$_3$ when the magnetic field is arranged perpendicular to the conducting planes ($\theta = 0°$) [7, 8, 11–15, 18]. A high-frequency oscillation (denoted as F_3 in the following) is observed with a strongly field-dependent amplitude (the double-peak structure of the F_3 oscillations is caused by Zeeman spin splitting). At low magnetic fields (see inset) as well as in the envelope of the signal at high magnetic fields, a lower-frequency oscillation can be recognized (called F_2 subsequently). For all κ-phase materials the Fermi surface (FS) is expected to consist of two extreme areas (see inset in Fig. 1). One of them is a closed lens-shaped orbit around *Z*, which encircles the extreme area A_2 and corresponds to the QO frequency $F_2 = 570$ T. The second one, a circular orbit covering A_3, corresponds very well to $F_3 = 3883$ T but is only closed as the gap ($E_g < 3$ meV) between *V* and *Z* is overcome by the so-called magnetic breakdown (MB) at sufficiently high magnetic fields (e.g., $B_{MB} \gtrsim 2$ T). For the carriers contributing to F_2 and F_3, respectively, the effective masses $m_2^* = 1.90m_e$ and $m_3^* = 3.90m_e$ (m_e = free electron mass) were obtained from the temperature dependence of the oscillation amplitudes [11, 12, 14–16, 18] by using the standard Lifshitz–Kosevich (LK) theory for QOs in metals [17].

During the investigations of κ-(BEDT–TTF)$_2$I$_3$ single crystals by QO experiments it turned out that regardless of the strong two-dimensionality of its electronic system,

the standard LK description for QOs in 3D metals [17] applies very well. Minor deviations from the LK behaviour can be understood by considering oscillations of the chemical potential (μ) with the QO frequency F_3 [14, 18]. This quite normal LK behaviour has been observed in the entire field, temperature and angular ranges covered by the experiments, except for a set of very specific experimental conditions, namely high magnetic fields (> 12 T), low temperatures and the special field orientation **B** ∥ (*b*,*c*) (i.e., $\theta = 0°$). There, strong deviations from LK behaviour are observed, which manifest themselves as a dramatic reduction of the SdH amplitudes.

This strong departure from the LK behaviour is demonstrated in Fig. 2 which shows the temperature dependence of the SdH amplitudes of F_2 and F_3 at 25 T and $\theta = 0°$, as obtained by the Fourier transformation. The dashed lines recall the expected standard LK behaviour, whereas the full lines are a guide to the eye to illustrate the damping effects. These damping effects influence (or may even prohibit) an estimation of the effective masses m^* under these special conditions [7, 8, 11, 12, 14–16, 18]. It should be recalled here that in contrast to the results shown in Fig. 2, experiments done at $\theta > 1°$ result in quite normal LK behaviour. The damping effects at $\theta = 0°$ were attributed [7, 8, 12]* to a localisation of electrons as possible in 2D electronic systems at low filling factors ν [13], where $\nu \equiv 1$ is known as the quantum limit (QL).

Since even at 52 T, which is the highest field applied in the investigations described above, the filling factors of both F_2 and F_3 are far away from QL [19], we searched especially for low-frequency quantum oscillations, although no small orbit on the FS corresponding to such a frequency is known from extended Hückel band structure calculations [4]. The search for such low-frequency oscillations requires QO experiments the be extended to the lowest possible fields, since the widest possible field windows in [1/B] are needed for their identification. Such low-field experiments are enabled for κ-(BEDT–TTF)$_2$I$_3$ by its high crystal quality, and they were performed as SdH experiments, since at low *B* this method is by far more sensitive than the dHvA torque technique. Figure 3a shows a low-field SdH experiment at $\theta = 0°$ and 0.38 K, where the signal is plotted after division by the background resistivity. The variation in the depth of the oscillations suggests a main frequency, corresponding to $F_0 = 13.2$ T, and its spin-split oscillations. It was shown in Refs. [7, 14, 18] that the 00 with F_0 corresponds to a very small pocket in *k*-space, whose area A_{F0} represents only about 0.3% of the FS. In view of this, it is not very surprising that this small closed orbit might be beyond the resolution of standard band structure calculations.

F_0 is observed at 0.4 K, above about 1.25 T. At 2 T, the SdH amplitudes of F_0 reach about 0.04% of the background resistivity. Above about 2.4 T, the amplitudes of F_0 could not be observed directly, since the amplitudes of F_2 and F_3 increase strongly

*It is widely discussed in Refs. [7, 8, 12, 19] why $\theta = 0°$ is the exclusive field orientation in a metallic multilayer 2DES, where two-dimensionality (and its results) may take effect. Therefore, this discussion is not given here.

with field and dominate those of F_0 (see Fig. 3a). The *action* of F_0 onto the amplitudes of F_2 and F_3, however, can be unambiguously observed up to high fields. This is illustrated in Fig. 3b by means of the so-called "Dingle plots" (DPs), given by the implicit

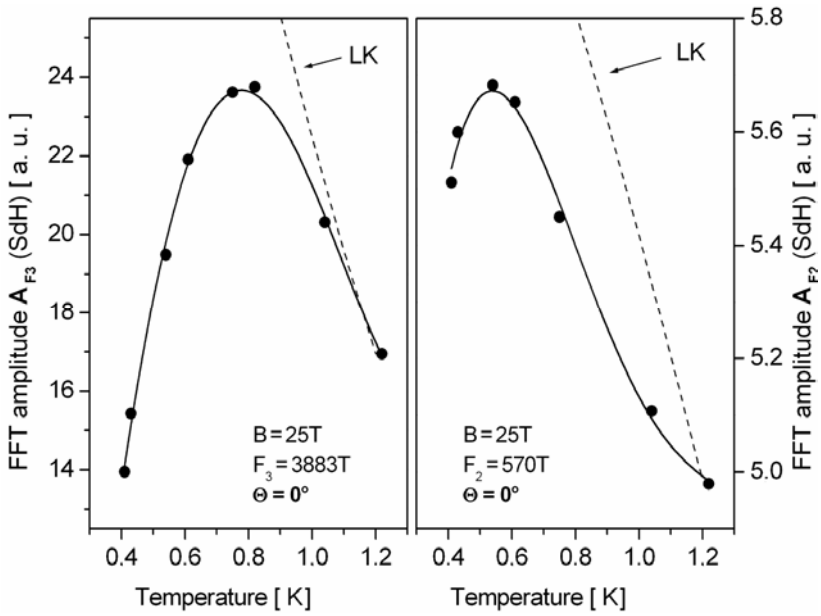

Fig. 2. Temperature dependence of the SdH amplitudes of F_2 and F_3 at 25 T and $\theta = 0°$ for κ-(BEDT–TTF)$_2$I$_3$. The dashed lines show the expected behaviour, according to the standard Lifshitz–Kosevich theory, whereas the full lines are guides to the eye

values of the FFT amplitudes of F_2 and F_3 (see left *y*-axis) versus 1/*B*. In the standard LK theory, a DP should be linear and its slope is a measure for the Dingle temperature T_D and the corresponding scattering time τ. In the present case of magnetic breakdown (MB), the LK theory has to be extended by the so-called "coupled network description" (CND), in order to account for the magnetic field dependence of the MB probabilities. In this case, the Dingle plot of F_2 should be sublinear, while that of the MB orbit F_3 is expected to remain linear with a modified slope (both curves are determined by the Dingle temperatures (T_D) and by the magnetic breakdown field B_{MB}). Even though the low field region is least influenced by the anomalous damping effects, it can only hardly be fitted by taking an exaggerated $B_{MB} \sim 4$ T and a far too high $T_D \sim 0.4$ K (see the dotted curve in Fig. 3b for F_2 and the dashed line for F_3). This indicates that already at low fields the behaviour of the QO amplitudes can hardly be described by the LK theory and the CND. At higher fields, the discrepancy becomes much stronger. Above 2 T, the DP of F_2 strongly deviates from the estimated curve (note the logarithmic scale). The DP of F_3 shows strong deviations from linearity above about 4 T. At high fields, both DPs show strong damping effects (grey-shaded areas), which cannot be explained by MB.

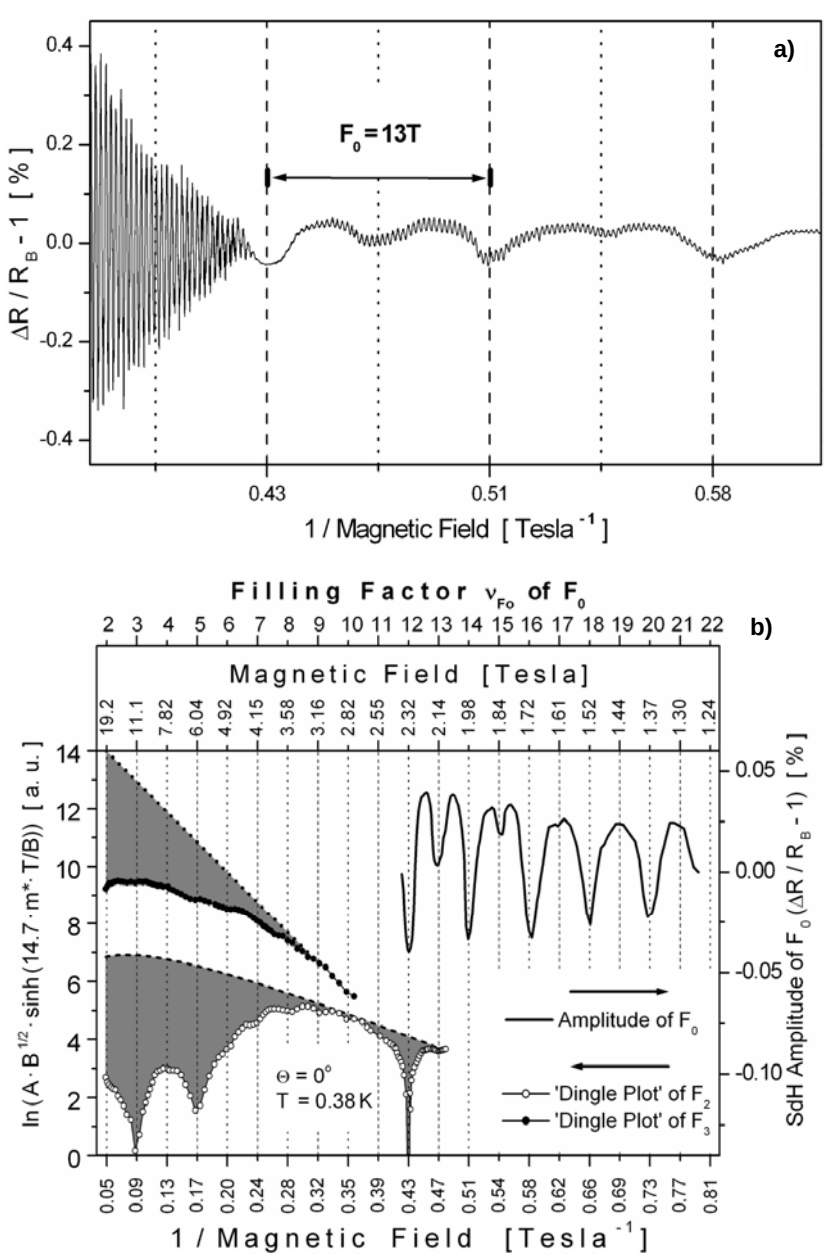

Fig. 3. Low-field SdH signal of a κ-(BEDT–TTF)$_2$I$_3$ single crystal versus 1/B at 0.38 K and θ = 0° after division by the background resistivity (a). SdH oscillations of F_0 in κ-(BEDT–TTF)$_2$I$_3$ single crystals at 0°. The filling factor ν of F_0 is indicated on the top axis (b), right). Dingle plots of F_2 and F_3 at 0° (see text) (b), left). The estimated dashed and dotted curves represent the expected amplitudes (accounting for magnetic breakdown). The grey-shaded areas represent the magnitude of the damping effects

The most important features in the DPs of both F_2 and F_3 are revealed by a "fine structure" of the DPs. These are discontinuities in their curvature at high fields. For understanding them, the field positions of the minima in the SdH signal of F_0 (where the Fermi energy E_F lies just between two successive spin-splitted Landau levels) are marked by grid lines and continued to high fields. The discontinuities and minima in the DP of F_2 show the same oscillatory structure as the oscillations with F_0. They occur just at the field values where the resistance minima of the F_0 oscillations are expected (i.e., where E_F lies between two adjacent spin levels of F_0, hence the corresponding ν is an integer; see top axis of Fig. 3b). The same behaviour (though weaker in magnitude) is present in the DP of F_3. Such an oscillatory structure with F_0 was observed in SdH experiments at $\theta = 0°$ on several crystals. By this, F_0 is identified to be directly involved in the damping of the amplitudes of F_2 and F_3. The filling factor ν_{F0} of F_0, as indicated on the top axis, turns out to be a controlling parameter for these effects [7]. While the amplitude of F_2 is already damped at $\nu_{F0} < 13$, that of F_3 is first demonstrably damped in the MB regime for $\nu_{F0} < 7$. The magnitude of the damping effects increases strongly with decreasing ν_{F0}. At the highest field applied, $\nu_{F0} = 2$ is reached with only two spin levels of the lowest Landau level of F_0 being occupied below the FS. While F_2 and F_3 are still at fairly high filling factors when the damping effects in their amplitudes occur, F_0 is at low ν already close to QL (the special situations at inverse field values $B^{-1} \sim 0.43$; 0.17 and 0.09 are discussed later). The observation illustrated with Fig. 3b – that the damping effects of the QO amplitudes of both F_2 and F_3 show an oscillatory pattern just with F_0 – proves that the electrons of all the corresponding "subsystems" (i.e., electronic bands) are strongly correlated. This correlation persists even though the involved bands are at very different filling factors ν. This established correlation means that the carriers contributing to F_2 and F_3 are themselves sensitive to the conditions introduced by F_0. This makes it easier to understand why the filling factor ν_{F0} of F_0 becomes a controlling parameter of the *entire* system and why at high fields it is able to force quantum limit conditions on the entire correlated electronic system.

Two-dimensionality and the obvious presence of electron correlation (EC) bring forth questions about their consequences, namely electron localisation around integer and noninteger low filling factors ν_{F0}. A correlation involving the electrons of all orbits proves that the resulting localisation effects may, accordingly, involve all carriers, not only those on the low-ν_{F0} orbit. Based on this, the damping effects of the QO amplitudes of F_2 and F_3 may be understood at this stage as a reduction of the number of mobile carriers contributing to these QOs. This reduction is caused by localisation effects, generated in turn by the low filling factors of F_0 [7, 16]. However, before going into detail with the discussion of this point, further decisive results are needed.

One of the fundamental differences between well-known semiconducting two-dimensional electron systems (2DESs) and the 2D organic metal κ-(BEDT–TTF)$_2$I$_3$ is the fact that in the former all electrons follow one single orbit, whereas in the latter they move on various orbits corresponding to very different QO frequencies. This

condition can be excellently used to probe variations of the chemical potential μ with the successive QO frequencies [8]. The huge differences between the frequency values of F_0, F_2, and F_3 allow one to use the high-frequency QOs as a 'high-resolution' sensor, which probes the actual position of μ and its low-frequency variations, if present (see Refs. [8, 19] for details).

In order to investigate more thoroughly the behaviour of μ in κ-(BEDT–TTF)$_2$I$_3$, the successive field positions B_n were determined, at which the actually highest occupied n-th Landau cylinder (LC) of a certain frequency (here, e.g., F_3) passes the Fermi cylinder. The corresponding Landau level indices n_{F3} can be obtained from Landau quantisation [8, 19].

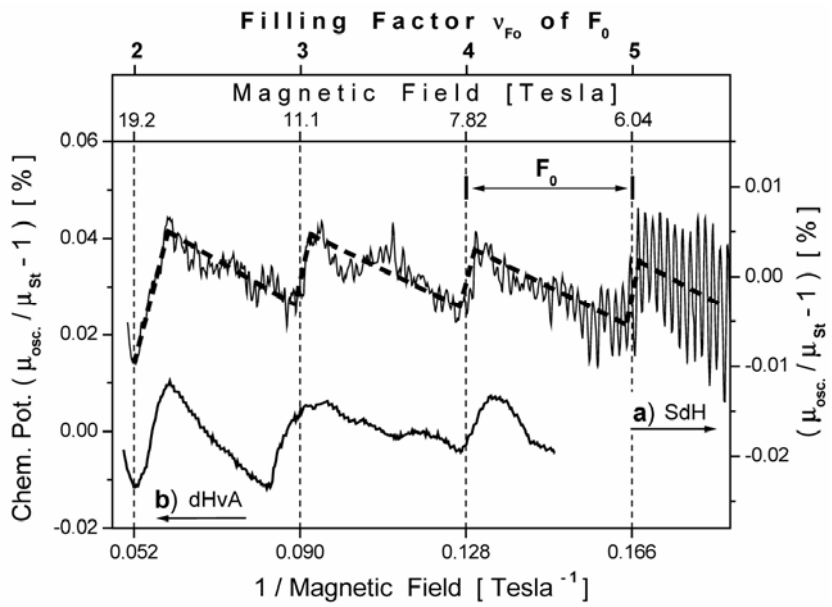

Fig. 4. Quantum oscillations of the chemical potential μ with F_0 at 0.38 K for κ-(BEDT–TTF)$_2$I$_3$. Curve a) was obtained from a SdH measurement at $\theta = 0°$ (see text), curve b) from a dHvA experiment at 9°, rescaled to 0° by the $1/\cos\theta$ law valid for 2EDSs ([8])

The result is plotted in Fig. 4. Curve a) is obtained from SdH measurements at $\theta = 0°$ and shows pronounced saw tooth oscillations of μ above c.a. 6 T, with an oscillatory sequence corresponding just to the low QO frequency $F_0 = 13$ T [16]. The filling factor ν_{Fo} and the corresponding magnetic field values are plotted on the top axes of Fig. 4. QOs with F_0 could not yet be detected above about 2.3 T (due to their low amplitude compared to the other QOs and background resistivity, see Fig. 3), whereas the μ oscillations with F_0, shown in Fig. 4, represent the detection of F_0 above 2.3 T up to high fields, where the concomitant damping effects in the SdH oscillations are prominent. This means that the μ oscillations with F_0 prove the presence of both a corresponding Landau cylinder structure and a corresponding closed orbit

on the FS. The fact that "sensors" (such as F_2 and F_3 oscillations) are able to probe the influence of F_0 confirms that the electrons on the F_0, F_2, and F_3 orbits are correlated. The very different filling factors of the "sensors" F_2 and F_3 and the correlated "object", namely F_0, proves that EC bridges the very different ν and that EC is operative even at very high values of ν of F_2 and F_3.

Let us now discuss in more detail the special situations at inverse field values B^{-1} ~ 0.43, 0.17 and 0.09, which are shown in Fig. 3. In the left part of the figure the Dingle plot of F_2 shows strongest damping effects at ν_{F0} = 3, 5 and 12. Even if they seem to be connected with the filling factor of F_0, they cannot be attributed in a conclusive way to this QO frequency, since they are far stronger than the damping effects at the neighbouring filling factors ν_{F0}. At first glance, the damping effects at ν_{F0} = 3 and 5 might be attributed to a strong spin polarisation and the proximity of the QL, but the strong amplitude reduction at the even ν_{F0} = 12 clearly contradicts both these possibilities. In Refs. [8, 19] it has been found that such effects cannot be explained by conventional damping mechanisms, even when going beyond those summarised in Ref. [17].

The strength of these special damping features in Fig. 3b gives rise to the question whether they may have a common origin beyond the existence of F_0. This in turn raises the question whether they are generated by a further low-frequency QO present in κ-(BEDT–TTF)$_2$I$_3$, whose lowest Landau level(s) only could be observed at high fields. The search for a further low-frequency oscillation in this material requires the SdH experiments to be extended further into the low-field range. This was enabled by decreasing T to dilution temperatures. It should be emphasised that this field region covers the magnetic breakdown between the closed F_2 orbit and open F_3 orbit. For this reason the oscillations of the F_3 orbit are a priori excluded in the investigations of the strong damping effects, since at these fields the F_3 orbit is not properly quantised and its investigation would be therefore influenced by MB effects.

Field sweeps were carried out on several κ-(BEDT–TTF)$_2$I$_3$ single crystals at 30 mK, with a very low sweeping rate and a field orientation perpendicular to the conducting planes (i.e., θ = 0°±0.04°). The results of these SdH experiments are summarized in Fig. 5. The low-field part of the detected DPs of several crystals in Fig. 5b show a new oscillatory structure with a frequency of 3.8 (±0.3) T, henceforth called F_{new}. The corresponding frequency is confirmed in the FFTs of the SdH signals [19]. They clearly show that the strongest damping effects are governed by the oscillatory structure with F_{new} alone. A number of arguments have been given to show that neither F_{new} = 3.8 T nor F_0 = 13 T can be generated by an assumed warping of the FS [19] and that this new oscillatory structure, with F_{new} = 3.8 T, has to be attributed to a new quantum oscillation (just as F_0). The corresponding extreme area $A_{F\text{new}}$ is very small, representing merely 10^{-3} of the first Brillouin zone. In view of this, it is not surprising that the small pocket corresponding to F_{new} has not yet been found by band structure calculations. In order to verify the presence of a Landau level structure corresponding to F_{new} by a thermodynamic property, the position of the chemical po-

tential μ and its variation was probed by the same method as before for the μ oscillations with F_0 [8, 19]. Due to the arguments discussed above, only the QOs with F_2 may be taken as "sensor" oscillations for probing μ. The results obtained on different crystals by SdH measurements at $\theta = 0°$ are depicted in Fig. 5a. The data show, with a very good agreement, an oscillatory structure with F_{new}. This confirms that F_{new} indeed corresponds to a thermodynamic quantity and confirms that F_{new} has to be identified with a quantised orbit on the FS and a Landau level structure.

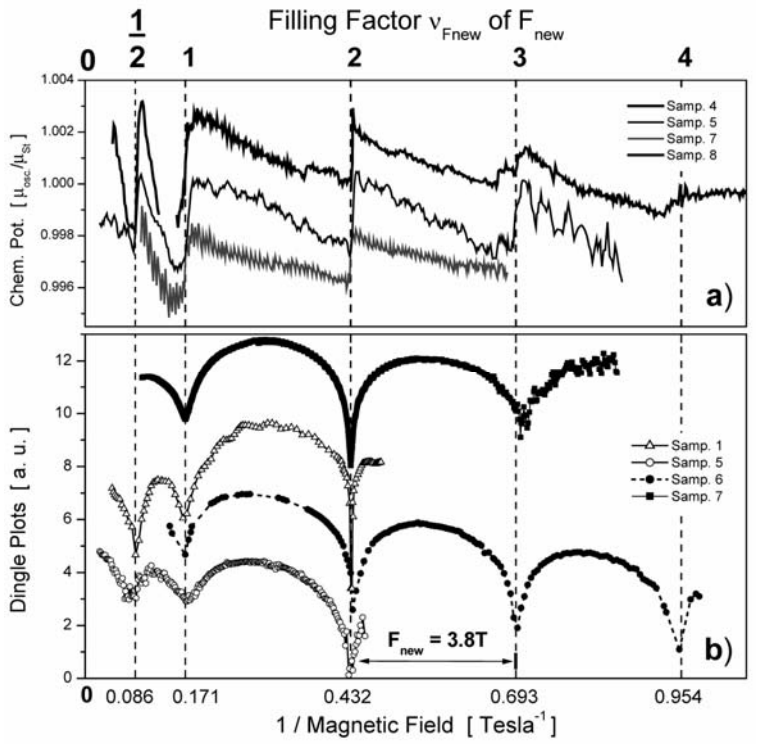

Fig. 5. Identification of the Landau level filling factors ν_{Fnew} of F_{new} in the 2D multilayer organic metal κ-(BEDT–TTF)$_2$I$_3$ at $\theta = 0°$: a) chemical potential oscillations detected on several single crystals versus ν_{Fnew}, b) Dingle plots of the F_2 amplitudes from several single crystals as obtained by SdH measurements at 0°. Note that the *x*-axes end up at a infinite magnetic field, namely $\nu \equiv 0$

The top axis of Fig. 5 shows the attribution of the Landau level filling factors ν_{Fnew} of F_{new} to corresponding chemical potential oscillations. Indeed, μ follows a saw tooth with its steep flank towards high fields and drops at integer values of ν, just as expected for an almost ideal 2DES. The quantum limit (QL), namely $\nu_{Fnew} = 1$, is reached at about 0.17 T^{-1}, or 5.9 T. The equidistant saw tooth track of ν_{Fnew} fits perfectly to the equidistant Landau level spacing of F_{new} between $\nu_{Fnew} = 1$ and 4.

At higher fields, i.e., at $\nu_{Fnew} < 1$, two fundamental conditions preset the interpretation of this extreme quantum limit region. First of all (as already mentioned),

$\nu_{F\text{new}} = 1$ is unambiguously identified, since no further assumed Landau level can be placed equidistantly at higher fields (on the left-hand side of $\nu_{F\text{new}} = 1$) in Fig. 5.

Secondly, the left margin of Fig. 5 is given by $1/B = 0$, i.e., an infinite field. This means that $\nu \sim 1/B \equiv 0$ at that point. These two definite conditions require that, within this extreme QL, the Landau level filling $\nu_{F\text{new}}$ be defined in rational parts of the remaining field intercept between $\nu_{F\text{new}} = 1$ and 0, which covers $\Delta[1/B] = [0\ \text{T}^{-1},\ 0.17\ \text{T}^{-1}]$. This reveals that the additional saw tooth oscillation at about $1/B = 0.086\ \text{T}^{-1}$ (i.e., $B = 11.6$ T) represents exactly $\nu_{F\text{new}} = 1/2$ in the multilayer 2D system. The fact that the thermodynamic property μ oscillates at $\nu_{F\text{new}} = 1/2$ proves the existence of a thermodynamically stable state. The presence of this state is also observed by strong damping effects in SdH oscillations at $\theta = 0°$ and $\nu_{F\text{new}} = 1/2$, as illustrated by the Dingle plots in Fig. 5b. Likewise, the integer $\nu_{F\text{new}} = 1$–4 can be identified in the Dingle plots as those field regions where the damping effects of SdH oscillations at 0° are strongest (note that strong damping effects are observed even at $\nu_{F\text{new}} = 4$, which is at about 1 T).

All in all, the most recent SdH experiments on κ-(BEDT–TTF)$_2$I$_3$ presented here reveal that low integer ν and even $\nu_{F\text{new}} = 1/2$ are present in this 2D multilayer organic metal. It is found that the values $\nu_{F\text{new}} = 1/2$; 1; 2; 3; 4 coincide with the positions of the strongest damping effects in SdH oscillations (with the anomalous minima in the Dingle plots) at 0° in this 2DES. On the one hand, theoretical descriptions are still lacking for such 2D multilayer organic metals with correlated carriers close to the QL. On the other hand, the experimental results were found to be in remarkable agreement with theoretical descriptions and experiments for semiconducting 2DESs, where electron correlation and localisation is considered. This agreement concerns a number of fundamental aspects, especially the observation of $\nu_{F\text{new}} = 1/2$; 1; 2; 3; 4 and their action onto the present correlated 2DES. It is straightforward that electron localisation in a 2DES around low ν may reduce the number of mobile carriers, thus leading to a damping of SdH amplitudes – a quantity which is only given by the (remaining) mobile carriers.

At a first glance, the observation of the $\nu = 1/2$ state in κ-(BEDT–TTF)$_2$I$_3$ is most exciting, especially since the originally expected single-layer fractional states of highest hierarchy [13], namely $\nu_{F\text{new}} = 1/3$; 2/3, ... are not observed here. On closer inspection, however, this is not too surprising in view of the fact that κ-(BEDT–TTF)$_2$I$_3$ is a *multilayer* 2DES. The present organic metal belongs undoubtedly to the category of coupled-multilayer systems with metallic*, finite interlayer tunnelling. In semiconducting multilayer 2DESs, $\nu = 1/2$ is expected under such conditions [22], and this filling fraction is indeed observed here in κ-(BEDT–TTF)$_2$I$_3$. Furthermore, for finite tunnelling, both odd and even integer ν are expected by theory [22] and are indeed

*In Ref. [20] it has been shown that interlayer transport in κ-(BEDT–TTF)$_2$I$_3$ is metallic, later in Ref. [21] it was specified that it is even coherent.

observed in the present metallic 2DES[*]. These are very important aspects, in which the results on κ-(BEDT–TTF)$_2$I$_3$ agree remarkably with the behaviour expected for semiconducting multilayer 2DESs.

A further view on the results of the preceding sections points out that the values $\nu_{F\mathrm{new}} = 1/2;\ 1;\ 2;\ 3;\ 4$ in the 2D multilayer organic metal κ-(BEDT–TTF)$_2$I$_3$ are brought into the system by 'subsystems'. These are the small pockets on the FS, which correspond to F_{new} and F_0, respectively, whereas the rest of the system, i.e., the orbits corresponding to F_2 and F_3, are still at fairly high ν. In view of this, the fact that low values of $\nu_{F\mathrm{new}}$ can be observed by their *action* on the electrons of the remaining system (i.e., those contributing to F_2 and F_3) proves the presence of sufficiently strong electron correlation to bridge over various bands with very different values of ν, thus involving even electrons with very high kinetic energy. Unfortunately, a theoretical description for such a complex situation in a correlated metallic multilayer 2DES is still lacking, and the electronic properties of κ-(BEDT–TTF)$_2$I$_3$ at high fields and low temperatures are still far from being understood.

3.2. Θ-(BEDT–TTF)$_2$I$_3$

Quantum oscillation experiments show two dominant oscillations with the frequencies $F_\alpha = 780$ T and $F_\beta = 4200$ T [10], which correspond very well to the proposed FS of Θ-(BEDT–TTF)$_2$I$_3$ [3–5]. This agreement is confirmed by angular magnetoresistance oscillations, which additionally show that the quasi-two-dimensional FS is warped, or corrugated [10]. In addition to F_α and F_β, a further very-low-frequency oscillatory structure was observed in magnetotransport with a frequency value between 2 T and 12 T [23], which cannot be attributed to the results of any of the band structure calculations quoted above. This oscillation (henceforth called F_γ) is observed up to 10 T, at which the filling factor $\nu_{F\gamma} = 1$ is reached [10]. A careful angular-dependent investigation showed that this oscillation is present not only for a field orientation perpendicular to the quasi-2D conducting planes, but even in a field orientation parallel to the planes. From these results it was concluded that the oscillatory structure can be attributed to a 3D pocket on the FS. Our SdH experiments clearly reproduce F_γ (comp. Ref. [10]). The presence of this very-low-frequency oscillation for fields orientated perpendicular and especially parallel to the conducting planes confirmed this 3D pocket on the FS. Attributing the F_γ oscillation to a real quantised orbit means that this Θ-phase salt indeed reaches the quantum limit at available fields. In view of this, and taking up the question of the strength of electronic two-dimensionality, we concentrated on the search for deviations in the temperature

[*] $\nu_{F\mathrm{new}} = 1/2$ and all further ν_{Fj} represent *total* filling factors of the *bulk* electronic system, *not*, e.g., the filling of a single layer. This corresponds perfectly to the case of coupled 2D (multi-) layers with interlayer tunnelling as described in Ref. [22].

dependence of SdH amplitudes, especially at $\theta = 0°$. This was verified, in order to find out whether this (quasi-)2D material might show similarly strong effects of two-dimensionality (i.e., damping effects of the QO amplitudes) as those observed for the κ-phase of (BEDT–TTF)$_2$I$_3$. With SdH measurements it was found that the temperature dependence of the oscillation amplitudes of both F_α and F_β show the same behaviour at low (9 T) and high fields (23 T) [10]. Consequently, the values $m^*_\alpha(9\text{ T}) = m^*_\alpha(23\text{ T})$ and $m^*_\beta(9\text{ T}) = m^*_\beta(23\text{ T})$ were obtained for each of these frequencies. The same behaviour was found for other tilt angles between the field and conducting planes, namely $\theta \neq 0°$. Hence, contrary to the properties of the 2D κ-phase salt, in which this tilt angle plays a decisive role, no difference in the behaviour of the SdH oscillations in the Θ-phase were found for $\theta = 0°$ and $\theta \neq 0°$. This means that despite its presumably low filling factors, the SdH amplitudes of the Θ-phase of (BEDT–TTF)$_2$I$_3$ do not show strong field dependent damping effects, as does κ-(BEDT–TTF)$_2$I$_3$ at $\theta = 0°$. This, however, is not surprising in view of the fact that the small pocket on the FS of the Θ-phase salt is a 3D one. The presence of this 3D pocket on the FS certainly influences the (quasi-)2D electronic behaviour of this material.

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