PDF hosted at the Radboud Repository of the Radboud University Nijmegen

The following full text is a publisher's version.

For additional information about this publication click this link.
http://hdl.handle.net/2066/29552

Please be advised that this information was generated on 2018-02-24 and may be subject to change.
Oscillatory magnetoresistance in the charge-transfer salt $\beta''$-(BEDT-TTF)$_2$AuBr$_2$ in magnetic fields up to 60 T: Evidence for field-induced Fermi-surface reconstruction

A. A. House
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

N. Harrison
Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Heverlee, Belgium

S. J. Blundell
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

I. Deckers
Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Heverlee, Belgium

J. Singleton
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

F. Herlach
Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Heverlee, Belgium

W. Hayes
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

J. A. A. J. Perenboom
Laboratorium voor Hoge Magneetvelden, Katholieke Universiteit Nijmegen, Toernooiveld 1, NL 6525 ED Nijmegen, The Netherlands

M. Kurmoo
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

P. Day
The Royal Institution, 21 Albemarle Street, London W1X 4BS, United Kingdom

(Received 6 October 1995)

Magnetoresistance measurements carried out in pulsed magnetic fields of up to 60 T and at temperatures down to 350 mK and angle-dependent magnetoresistance experiments performed in quasi-static fields have been used to establish that the charge-transfer salt $\beta''$-(BEDT-TTF)$_2$AuBr$_2$ undergoes a change in electronic structure at $\sim$10 T. We propose that this arises due to a field-induced transition between two different spin-density-wave states. Furthermore, at the highest magnetic fields, both the background magnetoresistance and effective masses of the quasiparticles were found to increase, possibly as a result of an enhancement of the density of states. It is found that frequency mixing effects at very high magnetic fields, observed in the Fourier spectra of the magnetoresistance, cannot be explained by the Shoenberg magnetic interaction, but are instead probably caused by oscillations in the chemical potential which become important as the extreme quantum limit is approached.

I. INTRODUCTION

Pulsed magnetic fields of $\sim$50 T have recently proved to be a powerful tool for the study of charge-transfer salts of the ion BEDT-TTF.$^{1-5}$ Some of these salts are found to undergo transitions at high magnetic fields between spin-density-wave (SDW) and metallic regimes (Refs. 2–7 and references therein); in addition, the amplitudes of quantum oscillatory phenomena such as the Shubnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) effects are found to grow dramatically with increasing magnetic field and appear to deviate significantly from conventional Lifshitz-Kosevich behavior.$^{2,3,8}$ In this paper we have used magnetoresistance measurements in pulsed fields of up to 60 T and the observation of angle-dependent magnetoresistance oscillations in static fields to demonstrate that the charge-transfer salt $\beta''$-(BEDT-TTF)$_2$AuBr$_2$ undergoes a field-driven transition between two different SDW ground states.

$\beta''$-(BEDT-TTF)$_2$AuBr$_2$ is a particularly interesting material, exhibiting a variety of apparent magnetic phase transitions below 30 K.$^{9-11}$ Furthermore, the low-temperature magnetoresistance of typical samples possesses a complex dependence on field strength and orientation,$^9$ plus a rich spectrum of SdH and dHvA oscillations.$^{9-11}$ These have
carried out very limited number of studies of this material have been other calculations, see Fig. 1 of Ref. 9 because of the low yield of crystals of the,9 which concentrated on the field region below 30 T;~

proven difficult to reconcile with the various predicted Fermi
sections, which either consist of quasi-one-dimensional
sheets or a single closed Q2D pocket plus a Q1D
portion of the field orientation dependence of the amplitude of
the results of that work are summarized in Fig. 1. An exami-
nation of the lock-in amplifier was set to 10 ms so that real-time
frequencies in the magnetoresistance of up to 20 kHz could
be detected without any noticeable attenuation. This enabled
magnetoresistance oscillations to be measured on the falling
(i.e., lower \( dB/dt \)) side of the field pulse14 down to the low-
est magnetic fields. Fields of up to 60 T were applied to the
sample, while temperatures down to 350 mK were provided
by means of a plastic \(^3\)He insert. Owing to the relatively
large resistivity of this material and small sample size, eddy
current heating15 was not significant; this fact was established
by comparing magnetoresistance data recorded with different
field pulse heights.2 The cryostat was mechanically decoupled
from the pulsed magnet to minimize the effects of vi-
brrational noise.

The angle-dependent magnetoresistance oscillations were
measured for a third sample (C) in static fields of up to 17 T
provided by a Bitter magnet at Nijmegen. The sample was
placed in a cryostat, which allowed it to be rotated about two
perpendicular axes in situ with a precision of \( \pm 1^\circ \). Cooling
was provided by \(^4\)He exchange gas, allowing the tempera-
ture range between 4.2 and 1.5 K to be studied. The crystal
was aligned by measuring the polarized infrared reflectivity
at room temperature. Contacts were made in the same man-
ner as for the pulsed field experiments and the measurement
was performed using standard four-wire ac techniques (20–
130 Hz), with the current directed perpendicular to the ac
planes. The magnitude of the current was 20 \( \mu \)A throughout
the experiment.

II. EXPERIMENTAL DETAILS

The \( \beta'^-(BEDT-TTF)_2AuBr_2 \) samples used in these experi-
ments were distorted hexagonal black platelets with dimen-
sions of approximately 0.5\( \times \)0.5\( \times \)0.1 mm\(^3\); the sample
growth and preparation details are identical to those de-
scribed in Ref. 9.

Pulsed field magnetoresistance measurements were per-
formed on two separate samples (A and B) at the Leu-
ven high field facility.15 The samples were orientated with their
conducting (2D) \( ac \) planes perpendicular to the magnetic
field. Four-wire contacts were made to the samples using
platinum paint and 25-\( \mu \)m gold wire, giving typical two-
terminal contact resistances of \( \sim 50 \) \( \Omega \). All experiments were
performed with an alternating current of 10 \( \mu \)A at 250 kHz
applied perpendicular to the conducting \( ac \) planes (see Ref. 9
for a discussion concerning the choice of this current direc-
tion). The voltage signal was preampified and subsequently
detected using a fast lock-in amplifier. The integration time
on the lock-in amplifier was set to 10 ms so that real-time
frequencies in the magnetoresistance of up to 20 kHz could
be detected without any noticeable attenuation. This enabled
magnetoresistance oscillations to be measured on the falling
(i.e., lower \( dB/dt \)) side of the field pulse14 down to the low-
est magnetic fields. Fields of up to 60 T were applied to the
sample, while temperatures down to 350 mK were provided
by means of a plastic \(^3\)He insert. Owing to the relatively
large resistivity of this material and small sample size, eddy
current heating15 was not significant; this fact was established
by comparing magnetoresistance data recorded with different
field pulse heights.2 The cryostat was mechanically decoupled
from the pulsed magnet to minimize the effects of vi-
brrational noise.

The angle-dependent magnetoresistance oscillations were
measured for a third sample (C) in static fields of up to 17 T
provided by a Bitter magnet at Nijmegen. The sample was
placed in a cryostat, which allowed it to be rotated about two
perpendicular axes in situ with a precision of \( \pm 1^\circ \). Cooling
was provided by \(^4\)He exchange gas, allowing the tempera-
ture range between 4.2 and 1.5 K to be studied. The crystal
was aligned by measuring the polarized infrared reflectivity
at room temperature. Contacts were made in the same man-
ner as for the pulsed field experiments and the measurement
was performed using standard four-wire ac techniques (20–
130 Hz), with the current directed perpendicular to the ac
planes. The magnitude of the current was 20 \( \mu \)A throughout
the experiment.

FIG. 1. (a) Fourier spectrum of the SdH oscillations in \( \beta'^-(BEDT-TTF)_2AuBr_2 \) for a field sweep at 490 mK with the magnetic
field perpendicular to the \( ac \) crystallographic plane [from
Doporto et al. (Ref. 9)]. Note the peaks occurring at 40, 140, 180,
and 220 T. (b) Shapes and orientations of the three Fermi-surface
pockets proposed by Doporto et al. (Ref. 9) from observation of the
angular dependence of the SdH oscillation amplitudes. (c) Section
through the first Brillouin zone of \( \beta'^-(BEDT-TTF)_2AuBr_2 \) showing the calculated room-temperature Fermi surface of Mori et al. (Ref.
12). (d) Reconstructed Brillouin zone and Fermi surface proposed
by Doporto et al. (Ref. 9).
The supposed presence of a SDW in \( \beta' \)-\( \text{BEDT-TTF}_2 \text{AuBr}_2 \) (see Ref. 9 and references therein for a discussion of the experimental evidence for a SDW ground state) and the observation of drastic changes in magnetoresistance gradient invite immediate comparisons with \( \alpha-(\text{BEDT-TTF})_2 \text{Mg(SCN)}_4 \) (\( M = \text{K}, \text{Tl}, \text{Rb} \)). The latter materials are also thought to exhibit SDW ground states and show strong “kinks” in the magnetoresistance,\(^{1–4,7,15,16}\) commonly associated with the destruction of the SDW due to the applied field. Part of the evidence for this is drawn from the fact that below \( \sim 5 \) K the classical background magnetoresistances of

\[ P = \exp(a_0 + a_1/B + a_2/B^2 + \cdots) \]

are strongly temperature dependent at fields below the “kink” (i.e., in the SDW state), but virtually temperature independent at fields above it (i.e., in the metallic state) (see Refs. 1–4, 7, 15, and 16 and references therein). The fact that the low-temperature magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \) is strongly temperature dependent up to the highest available field [Fig. 2(a)] therefore suggests that this material continues to remain in a SDW state up to 60 T for the temperature range examined in this study.

We turn to the oscillatory (SdH) component of the magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \). Previous studies on this material have shown hysteresis in the magnetoresistance at fields below \( \sim 20 \) T.\(^9\) This effect was also observable in the present experiments, but for fields below \( \sim 15 \) T on the rising side of the pulse, the oscillatory component of the magnetoresistance suffered some degree of attenuation as a result of sampling problems of the lock-in amplifier during the region of high \( dB/dt \) at the start of each pulse.\(^2,14\) For this reason the analysis of the quantum oscillations is restricted to the falling side of the pulse.

For the purpose of the analysis, \( \rho(1/B) \), the oscillatory component of the magnetoresistance, was divided by the classical background magnetoresistance \( P(1/B) \) in reciprocal magnetic field space. \( P(1/B) \) was approximated to an exponential polynomial of the form

\[ P = \exp(a_0 + a_1/B + a_2/B^2 + \cdots) \]

for the appropriate removal of the background magnetoresistance [as demonstrated in Fig. 2(c)].

\[ \alpha-(\text{BEDT-TTF})_2 \text{Mg(SCN)}_4 \] (\( M = \text{K}, \text{Tl}, \text{Rb} \)) salts are strongly temperature dependent at fields below the “kink” (i.e., in the SDW state), but virtually temperature independent at fields above it (i.e., in the metallic state) (see Refs. 1–4, 7, 15, and 16 and references therein). The fact that the low-temperature magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \) is strongly temperature dependent up to the highest available field [Fig. 2(a)] therefore suggests that this material continues to remain in a SDW state up to 60 T for the temperature range examined in this study.

We turn to the oscillatory (SdH) component of the magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \). Previous studies on this material have shown hysteresis in the magnetoresistance at fields below \( \sim 20 \) T.\(^9\) This effect was also observable in the present experiments, but for fields below \( \sim 15 \) T on the rising side of the pulse, the oscillatory component of the magnetoresistance suffered some degree of attenuation as a result of sampling problems of the lock-in amplifier during the region of high \( dB/dt \) at the start of each pulse.\(^2,14\) For this reason the analysis of the quantum oscillations is restricted to the falling side of the pulse.

For the purpose of the analysis, \( \rho(1/B) \), the oscillatory component of the magnetoresistance, was divided by the classical background magnetoresistance \( P(1/B) \) in reciprocal magnetic field space. \( P(1/B) \) was approximated to an exponential polynomial of the form

\[ P = \exp(a_0 + a_1/B + a_2/B^2 + \cdots) \]

for the appropriate removal of the background magnetoresistance [as demonstrated in Fig. 2(c)].

\[ \alpha-(\text{BEDT-TTF})_2 \text{Mg(SCN)}_4 \] (\( M = \text{K}, \text{Tl}, \text{Rb} \)) salts are strongly temperature dependent at fields below the “kink” (i.e., in the SDW state), but virtually temperature independent at fields above it (i.e., in the metallic state) (see Refs. 1–4, 7, 15, and 16 and references therein). The fact that the low-temperature magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \) is strongly temperature dependent up to the highest available field [Fig. 2(a)] therefore suggests that this material continues to remain in a SDW state up to 60 T for the temperature range examined in this study.

We turn to the oscillatory (SdH) component of the magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \). Previous studies on this material have shown hysteresis in the magnetoresistance at fields below \( \sim 20 \) T.\(^9\) This effect was also observable in the present experiments, but for fields below \( \sim 15 \) T on the rising side of the pulse, the oscillatory component of the magnetoresistance suffered some degree of attenuation as a result of sampling problems of the lock-in amplifier during the region of high \( dB/dt \) at the start of each pulse.\(^2,14\) For this reason the analysis of the quantum oscillations is restricted to the falling side of the pulse.

For the purpose of the analysis, \( \rho(1/B) \), the oscillatory component of the magnetoresistance, was divided by the classical background magnetoresistance \( P(1/B) \) in reciprocal magnetic field space. \( P(1/B) \) was approximated to an exponential polynomial of the form

\[ P = \exp(a_0 + a_1/B + a_2/B^2 + \cdots) \]

for the appropriate removal of the background magnetoresistance [as demonstrated in Fig. 2(c)].

\[ \alpha-(\text{BEDT-TTF})_2 \text{Mg(SCN)}_4 \] (\( M = \text{K}, \text{Tl}, \text{Rb} \)) salts are strongly temperature dependent at fields below the “kink” (i.e., in the SDW state), but virtually temperature independent at fields above it (i.e., in the metallic state) (see Refs. 1–4, 7, 15, and 16 and references therein). The fact that the low-temperature magnetoresistance of \( \beta'-(\text{BEDT-TTF})_2 \text{AuBr}_2 \) is strongly temperature dependent up to the highest available field [Fig. 2(a)] therefore suggests that this material continues to remain in a SDW state up to 60 T for the temperature range examined in this study.
tization has been measured using standard techniques, the Fourier spectra have been found to be dominated by four principal frequencies, \( \sim 40, \sim 140, \sim 180, \) and \( \sim 220 \) T [see, e.g., Fig. 1(a)]. In Fig. 3 the previously known frequencies of 140 and 180 T appear to be split into separate frequencies of 133 and 145 T, and 175 and \( \sim 195 \) T, respectively. Additional features are also present at \( \sim 53 \) and 240 T, whereas the frequencies at \( \sim 77 \) and 108 T are almost certainly second harmonics. The feature at \( \sim 20 \) T is an artifact of the background polynomial subtraction. At higher temperatures, as is apparent in Fig. 3, the 195 T frequency drops strongly in amplitude to reveal a further frequency at \( \sim 185 \) T. While there appear to be many new features in this data, the frequencies at 38 and 218 T are in accord with values reported in previous studies.9,10

The presence of apparent additive relationships between the components of the Fourier spectra has previously been interpreted as originating from the Shoenberg magnetic interaction.9,11 It has been speculated that this effect might also account for some of the SdH frequencies observed in \( \alpha-(BEDT-TTF)_2KHg(SCN)_4 \).15 However, recent absolute amplitude measurements of the dHvA effect in \( \alpha-(BEDT-TTF)_2KHg(SCN)_4 \) at high magnetic fields have shown that the internal fields are too small by at least four orders of magnitude. As the SdH frequencies in \( \beta'-(BEDT-TTF)_2AuBr_2 \) are even lower than those of \( \alpha-(BEDT-TTF)_2KHg(SCN)_4 \) (cf. Ref. 2 and this work), the internal fields generated by the oscillatory magnetization will be much smaller in \( \beta'-(BEDT-TTF)_2AuBr_2 \). Considered in the framework of the Lifshitz-Kosevich model for the amplitude of dHvA oscillations in a metal,13 it is now clear that the magnetic interaction is not a significant effect in \( \beta'-(BEDT-TTF)_2AuBr_2 \).

Since magnetic interaction is ruled out as an explanation of the multiple frequencies in \( \beta'-(BEDT-TTF)_2AuBr_2 \), another explanation must be sought. Figure 4(a) shows the first differential of the magnetoresistance with respect to the magnetic field, obtained by numerical differentiation of raw data such as those in Fig. 2(a). In Fig. 4(b) this procedure has also been applied to magnetoresistance data recorded using a Bitter magnet by Dopporto et al. (Ref. 9). (c) Landau level index of the SdH oscillation minima plotted against reciprocal magnetic field. This figure shows the reciprocal field regions over which different SdH frequencies dominate the Fourier spectrum.

From Fig. 4(c) it is clear that in the region 0.017–0.1 T \( (\sim 10–60 \) T), a SdH frequency \( F_{\gamma} \sim 222 \) T dominates the oscillatory component of the magnetoresistance, interfering with weaker SdH oscillations of other frequencies. In the region 0.1–0.16 T \( (6.3–10 \) T), a frequency \( F_{\alpha} \sim 191 \) T is strongest, until finally in the field region 0.16–0.25 T \( (\sim 4–6.3 \) T) the magnetoresistance minima correspond to a frequency \( F_{\beta} \sim 143 \) T. A detailed Fourier analysis of the quantum oscillations over different field ranges reveals that the \( F_{\gamma} \sim 222 \) T frequency appears only above 10 T, while the \( F_{\alpha} \sim 191 \) T frequency (which dominates the magnetoresistance between 6 and 10 T) vanished above \( \sim 14 \) T.

The Fourier transforms in Figs. 5(a) (above 14 T) and 5(b) (below 10 T) also show very clearly that different sets of SdH frequencies are present at high and low magnetic fields. At fields below 10 T three frequencies \( F_{\alpha} \sim 38 \) T, \( F_{\beta} \sim 143 \) T, and \( F_{\gamma} \sim 191 \) T are prominent [cf. Figs. 5(b) and 4(c)], whereas above 14 T, \( F_{\alpha} \sim 38 \) T, \( F_{\beta} \sim 175 \) T, and \( F_{\gamma} \sim 222 \) T dominate [cf. Figs. 5(a) and 4(c)]. In the intermediate region between 10 and 14 T, a poorly resolved mixture of frequencies is observed [Fig. 5(a), inset].

When performing Fourier transforms it was found that the modulation of the data by the Hanning window led to the introduction of an extra low-frequency component to the resulting Fourier spectra. For transforms taken across the entire field range, this effect was negligible, but for analysis over smaller regions of reciprocal space, this artificial peak became more significant and led to distortion of the low-frequency \( F_{\alpha} \sim 38 \) T SdH peak, thus shifting it to a lower frequency value. It was necessary to retain the use of a window function in order to avoid spurious peaks at higher frequencies on the Fourier transform, but it should be noted that the values that have been derived for the \( F_{\alpha} \) frequency were obtained by performing a Fourier transformation in the absence of any apodization.

It is worth remarking that similar transforms to that in Fig. 5(b) were obtained by Uji et al. using a field-modulation method to enhance the oscillatory component of the magnetoresistance.10,17 The reason for the plethora of fre-
quencies in the Fourier transform of the complete field range
(Fig. 3) is now apparent; different SdH frequencies dominate
the oscillatory component of the magnetoresistance over dif-
ferent field ranges. In order to clarify the changes in Fermi-
surface topology which gives rise to this phenomenon, we
turn to the angle-dependent magnetoresistance oscillations of
\( \beta' \)-(BEDT-TTF)\(_2\)AuBr\(_2\).

B. Angle-dependent magnetoresistance oscillations

The technique of observing magnetoresistance as a
sample is rotated in a constant magnetic field has become a
powerful tool for deducing the Fermi-surface (FS) topology
of organic metals. Both Q1D and Q2D sections of the FS can
cause angle-dependent magnetoresistance oscillations (AMRO’s)
via two different mechanisms; it is the Q2D mecha-
nism which is thought to be important in \( \beta' \)-(BEDT-
TTF)\(_2\)AuBr\(_2\).\(^9\) AMRO peaks due to a warped Q2D Fermi
surface have been thoroughly treated in Ref. 18 and are con-
nected with the vanishing of the electronic group velocity
perpendicular to the 2D layers. The angles between the normal
to the 2D planes and the magnetic field, \( \theta_i \), at which the
maxima occur are given by

\[
b'k_i\tan(\theta_i) = \pi(i \pm \frac{1}{2}) + A(\varphi),
\]

where the signs + and - correspond to positive and negative
\( \theta_i \), respectively, \( b' \) is the effective interplane spacing, \( k_i \)
is the maximum Fermi wave-vector projection on the plane of
rotation of the field, and \( i = \pm 1, \pm 2, \ldots \).\(^18\) Here positive \( i \) cor-
respond to \( \theta_i > 0 \) and negative \( i \) to \( \theta_i < 0 \). The gradient of a
plot of \( \tan(\theta_i) \) against \( i \) may thus be used to find one of the
dimensions of the Fermi surface, and if the process is re-
peated for several planes of rotation, defined by an azimuthal
angle \( \varphi \), the complete Fermi surface can be mapped.\(^{18,19}\) \( \varphi \)
is defined to be zero in the direction perpendicular to the
crystallographic \( c \) axis in the \( ac \) plane. \( A(\varphi) \) is a function
of the plane of rotation of the field, determined by the incli-
nation of the plane of warping; hence, this may also be
found.\(^18\)

Figure 6(a) shows the magnetoresistance plotted as a
function of tilt angle \( \theta \). Note that most of the features in
the magnetoresistance do not shift as \( B \) is increased, but merely
grow in amplitude; this is characteristic of AMRO’s, which
depend on the classical trajectories of the electrons across the
Fermi surface.\(^{15,18,19}\) However, for \( B \geq 10 \) T a new series of
strong oscillations grows, indicating that the Fermi surface
has changed in form. Note that one set of AMRO’s is ob-
served close to 90° at all magnetic fields and has a period
\( \Delta(\tan(\theta)) \) of between 2 and 4, depending on the azimuthal
angle \( \varphi \) \( \left[ \text{Fig. 6(b)} \right] \); such a large value indicates a small
Fermi-surface pocket, which we shall label the \( \alpha \) pocket. On
the other hand, the AMRO’s which appear for fields above
10 T \( \left[ \text{Fig. 6(a)} \right] \) have a smaller period in \( \tan \theta \), indicating that
they are associated with a larger area Q2D pocket. Their
appearance at the same field as the \( F_{y} \approx 222 \) T frequency
SdH oscillations \( \left[ \text{see Figs. 4(a) and 5(a)} \right] \) strongly suggests
that the two phenomena are associated with the same Fermi-
surface pocket. We shall refer to this section of Fermi surface
as the \( \gamma' \) pocket.

Assuming for simplicity that the Fermi-surface pockets
involved possess elliptical cross sections, the projection vec-
tor \( k_i \) (normal to the tangent) is related to the radii of the
major semi axis \( k_x \) and minor semiaxis \( k_y \) of a particular ellipse
by

\[
k_i = [k_x^2 \cos^2 (\varphi - \xi) + k_y^2 \sin^2 (\varphi - \xi)]^{1/2},
\]

where \( \varphi \) is again (see above) the azimuthal angle describing
angular position in the plane of the ellipse and \( \xi \) is the incli-
nation of the major axis with respect to \( \varphi = 0 \). For an elong-
ated ellipse \( \text{Eq. (2)} \) describes a locus in the form of a figure
8 in polar \( k_i \varphi \) space, as shown in Fig. 7(a).

Figure 7(b) shows a polar plot of \( k_i \) derived from the
periodicity of the AMRO’s close to \( \theta = 0 \) in Fig. 6(b), to
which \( \text{Eq. (2)} \) has been fitted, plus the shape of the derived
elliptical Fermi-surface cross section. The AMRO’s which emerge above \( \sim 10 \) T indicate that the \( \gamma' \) pocket \( \left( F_{y} \approx 222 \right. \) T
SdH frequency) is very elongated, with its long axis perpen-
dicular to the crystallographic \( c \) direction. The limited azi-
muthal resolution of the AMRO experiments means that the
length of the short axis \( k_y \) cannot be determined to any rea-
nable accuracy; furthermore, the width determined for this
elongated section of Fermi surface will also be prone to error
if the pocket is not perfectly elliptical. For these reasons,
the AMRO data were fitted using \( k_x \) and \( \xi \) as adjustable para-
meters, but with the ellipse area constrained to correspond to a
SdH frequency of 222 T. This fit is presented in Fig. 7(b) and
yields values of \( k_x = (3.42 \pm 0.07) \times 10^9 \) m\(^{-1} \) and \( k_y = 1.6 \times 10^8 \)
m\(^{-1} \). The total length of the major axis of the ellipse, \( 2k_x \),
exceeds the width \( (7.28 \times 10^9 \) m\(^{-1} \)) of the room-temperature
Brillouin zone \( \left[ \text{Fig. 1(c)} \right] \). Since the major axis of this ellip-
tical pocket is inclined at \( (12 \pm 2) \)° to the reciprocal lattice.
vector $a^*$ (which lies along the direction of the Brillouin zone width), this pocket takes the form shown in Fig. 8.

The AMRO’s which occur closer to 90°, both above and below 10 T, can be fitted by a more isotropic 2D Fermi-surface section, as shown in Figs. 7(c) and 7(d); these figures are derived from AMRO data recorded at 5 and 17 T, respectively, illustrating the field independence of this pocket. The
accuracy of the length of the minor axis of a more isotropic Fermi-surface pocket does not depend so much on a high azimuthal resolution. Hence the value obtained for both axis parameters \( k_x \) and \( k_y \) is much more reliable. From the fit to the 17-T data, we get \( k_x = (5.73 \pm 0.1) \times 10^8 \) m\(^{-1}\), \( k_y = (2.49 \pm 0.4) \times 10^8 \) m\(^{-1}\), and \( \xi = -(21 \pm 4) \), implying that the minor axis of the pocket is rotated by this angle from the crystallographic \( c \) axis. The values of \( k_x \) and \( k_y \) indicate that the area of this Fermi-surface pocket is \( S_a = (4.48 \pm 0.7) \times 10^{17} \) m\(^2\), corresponding to a SdH frequency of 47\( \pm 8 \) T. A fit to the 5-T data yields \( k_x = (6.04 \pm 0.3) \times 10^8 \) m\(^{-1}\), \( k_y = (2.66 \pm 0.7) \times 10^8 \) m\(^{-1}\), and \( \xi = -(24 \pm 7) \), giving \( S_a = (5.05 \pm 1.3) \times 10^{17} \) m\(^2\), which corresponds to a SdH frequency of 53\( \pm 14 \) T. This strongly suggests that the \( \alpha \) pocket is largely unaffected by any field-induced Fermi-surface reconstruction and that it is associated with the \( F_{\alpha} \) and \( F_{\delta} \) SdH frequency present over all field ranges [see Figs. 3, 5(a), and 5(b)].

Support for the above assignments of AMRO features to particular Fermi-surface pockets may be derived from data presented in Ref. 9. The geometrical considerations associated with the observation of AMRO’s are also thought to lead to oscillations in the amplitudes of the SdH frequencies9,18 and this was the technique used in Ref. 9 to derive the approximate Fermi-surface shapes illustrated in Fig. 1(b). Although this method is time consuming and of very limited accuracy [the SdH frequencies disappear at relatively low values of tan(\( \theta \)) (Ref. 9)], it can potentially lead to a direct association of the Shubnikov–de Haas oscillation frequency with a particular shape of Fermi surface. If one compares Fig. 6 of Ref. 9 with Figs. 7(b)–7(d) of this work, it is apparent that the Fourier amplitude of the 220 T SdH frequency (i.e., \( F_{\gamma'} \) in this work) oscillates at small values of \( \theta \) in a similar manner to the AMRO’s which occur only at fields above 10 T whereas the amplitude of the 40 T SdH oscillations (i.e., the \( F_\alpha \) series) only begins to oscillate with tilt angle at much higher \( \theta \).

However, the limited number of azimuthal angles studied by Doperto et al.9 means that the derived Fermi-surface shapes shown in Fig. 1(b) are only very rough guides; this is especially true for Fermi-surface pockets which are very anisotropic in shape. As a result, Doperto et al. obtained areas derived from the angle dependence of the SdH oscillations which they recognized to be too large, and they took this to imply that the SDW nesting vector included an interplane component.9 The present work illustrates that if AMRO’s are recorded at the large number of azimuthal angles necessary to obtain accurate Fermi-surface shapes, then the derived pocket areas agree with the SdH frequencies to within experimental errors; hence, we find that there is no evidence for an interplane component of the SDW nesting vector.

Additional complications affect the method used to derive the Fermi-surface pocket shapes in Ref. 9. The amplitudes of the SdH oscillations of the \( \gamma' \) pocket could have been affected by the background magnetoresistance, which was not taken into account; furthermore, the strong AMRO’s of this pocket could partly modulate the SdH oscillations of the other pockets. Other effects, such as a spin zero in the SdH oscillations,13 could also cause problems by leading to extra minima. It can therefore be concluded that the shapes of the \( \alpha \) and \( \gamma' \) Fermi-surface pockets derived in this work are much more reliable than the previous estimates shown in Fig. 1(b).9

IV. DISCUSSION

A. Field-induced Fermi-surface changes

The data reviewed in the previous section provide evidence that the Fermi surface of \( \beta'-(BEDT-TTF)_2\text{AuBr}_4 \) undergoes a field-induced change in form between \( \sim 10 \) and \( \sim 14 \) T. At fields below 10 T the Fermi surface appears to be characterized by three small Q2D pockets, which we refer to as \( \alpha, \beta, \) and \( \gamma \), with areas corresponding to the SdH frequencies \( F_{\alpha} \approx 38 \) T, \( F_{\beta} \approx 143 \) T, and \( F_{\gamma} \approx 191 \) T, respectively. There was speculation in Ref. 9 as to whether the \( F_{\beta} \) frequency corresponds to a real carrier pocket or whether it results from some kind of frequency mixing effect. However, as is seen in Fig. 4(c), the \( F_{\beta} \) frequency dominates the SdH oscillations at fields below 6 T, implying that it must be a real Fermi-surface pocket.

At fields above 10 T the \( F_{\alpha} \) frequency persists (as do the associated AMRO’s), while the \( F_{\beta} \) and \( F_{\gamma} \) frequencies vanish, to be replaced at fields above \( \sim 14 \) T by two new frequencies \( F_{\beta'} \approx 175 \) T and \( F_{\gamma'} \approx 222 \) T; we refer to the associated Fermi-surface pockets as \( \beta' \) and \( \gamma' \), respectively. The AMRO measurements indicate that the \( \gamma' \) pocket is a very elongated ellipse; however, neither the \( \beta \) or \( \gamma \) pockets in the low-field (<10 T) state nor the \( \beta' \) pocket in the high-field state (\( B > \sim 14 \) T) results in strong AMRO’s. A number of weak features away from the strong \( \alpha \) and \( \gamma' \) AMRO’s may be perceived in Figs. 6(a) and 6(b), but these were too indistinct for quantitative analysis. The reasons for the nonappearance of Q2D AMRO’s have been recently discussed in Ref. 19; it is likely that the \( \beta, \gamma, \) and \( \beta' \) pockets are too irregularly or severely corrugated to produce oscillations, as the Q2D AMRO effect relies on weak and regular corrugation of the section of Fermi surface in question.

It is instructive to compare the \( \alpha \) and \( \gamma' \) Fermi-surface pockets measured in the high-field state (i.e., deduced from the AMRO’s) with the Fermi-surface calculation of Mori et al.12 (Fig. 8). Given the shape of the \( \alpha \) pocket and also the fact that it is not affected by any rearrangement of the Fermi surface, we can associate it with the Q2D section predicted by Mori et al., centered on the X point of the Brillouin zone. The size of this pocket is much smaller than the bandstructure calculation predictions (derived from room-temperature crystallographic data), implying that a determi-
nation of the low temperature crystal structure and improved calculations are required. On the other hand, the elongated \( \gamma' \) pocket is probably generated by the imperfect nesting of the Q1D portions of the Fermi surface. Imperfect nesting should result in at least two pockets and so is likely to be responsible for the \( \beta'' \) pocket, which should therefore lie roughly in the same plane as the \( \gamma' \) pocket. From the charge transfer known to occur in the \( \beta'' \) pocket, it is expected that the band structure should be exactly three-quarters filled.\(^9\)\(^–\)\(^12\) For this reason \( \beta'' \)-(BEDT-TTF)\(_2\)AuBr\(_2\) is a compensated semimetallic compound in which the areas corresponding to the occupied and unoccupied states in the first Brillouin zone should be equal. As a result of the SDW, parts of the open pieces of the Fermi surface disappear and the total areas of remaining hole and electron orbits should be equivalent, explaining the approximate additive relationships between the various SdH frequencies (i.e., \( F_a + F_{\beta''} \approx F_{\gamma'} \)). Taking into account the above consideration for the high-field region in \( \beta'' \)-(BEDT-TTF)\(_2\)AuBr\(_2\), we should expect the \( \gamma' \) frequency to be an electron pocket, roughly compensating in area for the \( \alpha \) and \( \beta' \) hole pockets. Similar considerations apply in the low-field state below 10 T where the \( \beta \) and \( \gamma \) pockets will result from the imperfect nesting of the Q1D Fermi-surface sections and \( F_a + F_{\beta''} \approx F_{\gamma'} \); in this case, \( \gamma \) is expected to be the electron pocket.

We have now provided an interpretation of the abundance of SdH frequencies present in the Fourier transform covering the entire field range (2–60 T); some correspond to the “low-field” (<10 T) Fermi surface, whereas others are due to the “high-field” (>10–14 T) Fermi surface. Only the 53 and 133 T frequencies observed in Fig. 3 have not been accounted for. One possible explanation of these extra frequencies is that they correspond to the maximum (belly) cross section of the \( \alpha \) pocket and the minimum (neck) cross section of the \( \beta \) pocket, respectively, resulting from the warping of the Fermi surface in the reciprocal lattice \( k_x \) direction; by inspection of Fig. 3, it appears that the frequencies are paired as 38 T with 53 T and 133 T with 145 T. However, if such an interpretation were true, the same splitting should be observable over any magnetic field range provided that the interval in \( 1/B \) space was sufficiently long to give the Fourier transform an adequate resolution.\(^13\) The lowest magnetic field studies have been performed by Uji \textit{et al.}\(^10\) whose experiments covered the field interval from 2.25 to 13.8 T; this corresponds to a \( \sim0.37 \) \( T^{-1} \) range in \( 1/B \), approximately twice that in our pulsed field data. The absence of any splitting of the frequencies in the data of Uji \textit{et al.} suggests two things: first, that the warping of the Fermi surface is not a significant factor in \( \beta'' \)-(BEDT-TTF)\(_2\)AuBr\(_2\) and, second, that the apparent splitting of the frequencies is simply an artifact of extending the measurements to very high magnetic fields, probably resulting from the changes in the Fermi surface occurring between 10 and 14 T. The Fourier transform of the region between 10 and 14 T (Fig. 5(a), inset) exhibits a poorly resolved mixture of frequencies, including one close to 133 T. The “split” frequencies are therefore perhaps characteristic of this intermediate transition region between the high-field (>14 T) and low-field (<10 T) regimes. Such an extended field width between two different states of a SDW is not without precedent in BEDT-TTF salts; the true width of the “kink” transition between the SDW and metallic states of \( \alpha \)-(BEDT-TTF)\(_2\)KHg(NCS)\(_4\) has recently been shown to be \( \sim7 \) T.\(^5\)

**B. Extreme quantum limit**

In magnetic fields of 60 T the \( \alpha \) frequency reaches the extreme quantum limit where the last Landau level is only partially occupied. The extent to which the last level is occupied will depend on the deviation of the chemical potential \( \mu \) from its zero-field value. The simple depopulation of a Fermi-surface pocket is usually expected to lead to a reduction in the scattering cross section of the quasiparticles and therefore to an overall drop in the magnetoresistance.\(^13,20\)\(^–\)\(^25\) The observed strong increase in the background magnetoresistance at high fields is thus contrary to the expectations of this simple model.

A number of recent theoretical papers\(^26\) have speculated that strong electron self-energy effects will occur as a Q2D metal approaches the extreme quantum limit. This is expected to lead to an enhancement in the density of states (DOS) at the Fermi energy and thus to promote phenomena such as reentrant superconductivity. Given that superconducting and SDW ground states are often found to be competing effects in organic conductors, we might expect the existence of a SDW in \( \beta'' \)-(BEDT-TTF)\(_2\)AuBr\(_2\) to prevent superconductivity from taking place. An increase in the DOS at high fields or, alternatively, a reduction of the energy separation of the states could be expected to increase the quasiparticle scattering cross section at high fields and perhaps account for the strong increase in the magnetoresistance which is observed experimentally.

Further evidence for the increase in the DOS at high magnetic fields is found in the values of the quasiparticle effective masses derived from the SdH oscillations over the range from 0.07 T\(^{-1} \) (<14 T) to 0.017 T\(^{-1} \) (<60 T).\(^{27,31}\) [Doporto \textit{et al.}] evaluated the effective masses in \( \beta'' \)-(BEDT-TTF)\(_2\)AuBr\(_2\) at magnetic fields \( \leq17 \) T, where hysteresis complicates the required analysis of the magnetoresistance; we avoid this problem by evaluating the effective masses at higher magnetic fields.] The standard procedure\(^13\) for extracting the oscillatory component of the magnetoresistance by dividing by the background magnetoresistance \( P(1/B) \) was followed, and the temperature dependence of the Fourier amplitude \( a \) of the SdH oscillations was fitted to the equation\(^2,13\)

\[
a=a_0x/\sinh x, \tag{3}
\]

where

\[
X=2\pi^2 p k T / h \omega_c \sim 14.69 m \* T \, / \, B.
\]

Table I shows the derived effective masses, which are noticeably higher than the low field (i.e., \( \lesssim 17 \) T) estimates in Ref. 9. Large effective masses have also been observed in other organic conductors in high magnetic fields\(^2,8,27,31\) and have been attributed to a transition from quasi- to ideal two-dimensional behavior of the quasiparticles with increasing field.\(^8,27,31\) However, recent numerical simulations indicate that the latter effect is unlikely to be responsible for the experimental observations; even in the most extreme case of a zero scattering rate (\( \tau^{-1}=0 \)), the simulations reveal that only the mass estimates of the second and higher harmonics
of the quantum oscillations are strongly affected,\textsuperscript{28} in agreement with recent experimental data.\textsuperscript{2} The process of forcing a fit to the Lifshitz-Kosevich theory\textsuperscript{13} to the first harmonic amplitude, even for the most 2D material, does not seem to produce a large deviation in the effective mass estimate.\textsuperscript{28} The values and amplitudes of the frequencies observed cannot be due to magnetic breakdown.\textsuperscript{13,30} The sharp minima observed in $\beta'$-(BEDT-TTF)$_2$AuBr$_2$ therefore contrast with the simple behavior expected, and the reasons for this are as yet unclear.

### V. CONCLUSION

Single crystals of $\beta'$-(BEDT-TTF)$_2$AuBr$_2$ have been studied using pulsed magnetic fields of up to 60 T and angle-dependent magnetoresistance oscillation (AMRO) measurements carried out in quasi-static fields. The strong temperature dependence of the magnetoresistance of $\beta'$-(BEDT-TTF)$_2$AuBr$_2$ and the persistence of several Shubnikov–de Haas (SDH) frequencies at fields of up to 60 T indicate that the spin density wave (SDW) previously noted in this material\textsuperscript{9} persists to the highest available fields. Both the SDH oscillations in the magnetoresistance and the AMRO measurements show that two distinct phases, each with a characteristic Fermi surface, exist within this SDW state; these correspond to magnetic fields of $<10$ T and magnetic fields $>14$ T, respectively, and are separated by an apparently extended transition region between $10$ and $14$ T. The Fermi surface in the low-field regime consists of three pockets $\alpha$, $\beta$, and $\gamma$, with areas corresponding to the SDH frequencies $F_\alpha\approx 38$ T, $F_\beta\approx 143$ T, and $F_\gamma\approx 191$ T. Above 14 T the AMRO measurements show that the $\alpha$ pocket is unaltered, indicating that it probably corresponds to the closed quasi-two-dimensional (Q2D) Fermi-surface section in the calculations of Mori et al.\textsuperscript{12} In contrast, above 14 T the $\beta$ and $\gamma$ Fermi-surface sections are replaced by $\beta'$ and $\gamma'$ pockets, with areas corresponding to SDH frequencies $F_{\beta'}\approx 175$ T and $F_{\gamma'}\approx 222$ T. The $\beta$, $\gamma$, $\beta'$, and $\gamma'$ pockets probably result from imperfect nesting of Q1D sections of the high-temperature Fermi surface; support for this assignment is given by the AMRO measurements, which show that the $\gamma'$ pocket is very elongated with its long axis parallel to the crystallographic $a$ direction.

At magnetic fields approaching 60 T, a strong increase in the magnetoresistance and larger effective masses than those measured at low fields are observed, perhaps indicating that the density of states is enhanced as a result of the approach of the ultraquantum limit. The Landau levels in this field region appear to be very sharply defined so that mixing of the Shubnikov–de Haas frequencies occurs due to strong oscillations of the chemical potential.

### ACKNOWLEDGMENTS

This work is supported by EPSRC (UK), the Royal Society (UK), and the European Union HCM, TML, and Large Facility Programs. The Leuven Laboratory is supported by the National Fonds voor Wetenschappelijk Onderzoek. A.H. wishes to thank the Erasmus program for financial support. N.H. is supported by the Onderzoeksaad, K. U. Leuven. We should like to thank Professor G. Pitsi and Professor C. Agosta for provision of components for the $^3$He cryostat, and Dr. S. Uji for fruitful discussions.

---

### TABLE I. Effective masses in the high-field state.

<table>
<thead>
<tr>
<th>$F(T)$</th>
<th>Label</th>
<th>$m^*(m_e)$</th>
<th>$1/B = 0.017–0.1 \text{T}^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>38</td>
<td>$\alpha$</td>
<td>1.9(3)*</td>
<td></td>
</tr>
<tr>
<td>175</td>
<td>$\beta'$</td>
<td>4.5(2)</td>
<td></td>
</tr>
<tr>
<td>222</td>
<td>$\gamma'$</td>
<td>4.0(1)</td>
<td></td>
</tr>
<tr>
<td>370</td>
<td>$\beta' + \gamma' - \alpha$</td>
<td>3(1)</td>
<td></td>
</tr>
<tr>
<td>404</td>
<td>$\beta' + \gamma'$</td>
<td>4.7(5)</td>
<td></td>
</tr>
<tr>
<td>440</td>
<td>$2\gamma'$</td>
<td>4.4(4)</td>
<td></td>
</tr>
</tbody>
</table>
Magnetic Oscillations in Metals

1986


17. In the very-low-field studies of Uji et al. (Ref. 10), further higher frequencies have been observed in the Fourier spectra, which have been accounted for as sum and difference frequencies of the lower peaks present. It has been proposed that these frequencies result from the Shoenberg magnetic interaction, but in the light of recent evidence (Ref. 2), it is clear that an alternative explanation must be sought. Sum frequencies could result from magnetic breakdown or could possibly arise from the application of the field-modulation technique over a region of the magnetoresistance where it is known to be strongly hysteretic.


22. R. G. Clark (private communication).


