Superconducting Fluctuations and the Reduced Dimensionality of the Organic Superconductor $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ as Observed through Measurements of the de Haas-van Alphen Effect

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We report measurements of the de Haas-van Alphen (DHVA) effect and AC susceptibility in the layered organic superconductor $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. The amplitude of the DHVA oscillations is attenuated over a wide field range above the irreversibility line, $B_{irr}(B)$, below which a rigid flux lattice is formed. Thus the DHVA effect provides a unique probe of the superconducting fluctuations at high fields in this material. We compare our measurements with other determinations of the superconducting phase diagram of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$.

I. INTRODUCTION

$\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ is an organic superconductor consisting of alternating layers of conducting BEDT-TTF and insulating Cu(NCS)$_2$ anions. This complicated structure yields a simple Fermi surface consisting of quasi two-dimensional (Q2D) pockets, and open quasi one-dimensional (Q1D) sheets. The layered structure is reflected in the anisotropic properties of the superconductivity ($T_c \sim 9$ K), appearing in transport, magnetization, AC susceptibility, and torque magnetometry experiments. The low dimensionality together with short coherence length, result in fluctuations of the superconducting order parameter being important over a wide range in temperature and applied field. The importance of thermal fluctuations or quantum fluctuations leads to, for example, a broadened anomaly in the specific heat capacity. The superconducting phase diagram of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ has a complicated structure. A vortex liquid phase is thought to exist over a wide $B-T$ region bounded by the irreversibility line at low temperatures and fields.

Recent experience has shown that the de Haas-Alphen (DHVA) effect is a powerful probe for in the investigation of the mixed state of superconductors. The DHVA effect consists of an oscillatory variation of the magnetization $M$ which varies periodically in inverse applied field. For a two-dimensional metal, each Fermi surface sheet with area $A$, gives rise to a fundamental oscillatory magnetization,

$$
\vec{M} = \frac{e^2 VF}{bm_\pi} \frac{X}{\sinh X} \exp(-\pi m_b/eB\theta_0) 
\times R_s(B) \times \cos \left( \frac{2\pi F}{B} \theta + \phi \right), \tag{1}
$$

where $B$ is the applied field, $b$ is the thickness of the 2D layer, $V$ is the volume of the sample, $F = (h/2\pi e)A$, $m^*$ is the renormalised cyclotron mass, $m_b$ the band mass and $X = 2\pi m^* k_B T/eB$. DHVA oscillations in the superconducting mixed state were first observed in NbSe$_2$ and have now been measured in many superconductors, such as A15 materials, borocarbides, organic superconductors, etc., and in some heavy fermion superconductors. On entering the superconducting (mixed) state from high fields, the effect of the superconductivity is to cause an attenuation of the amplitude of the oscillatory magnetization which can be described by the factor $R_s$ in Eq. (1). In the normal state $R_s(B_0) = 1$.

Many theories have been developed to account for the DHVA effect in the mixed state, all of which implicate the superconducting energy gap, $\Delta$, in the signal attenuation. By performing angle resolved DHVA measurements, it should be possible to probe any anisotropy in $\Delta$, which is an issue of recent intensive debate in organic superconductors. There is considerable evidence that the $\Delta$ of the $\kappa$-type BEDT-TTF organic superconductors displays $d$-wave symmetry with line nodes. Evidence includes NMR susceptibility, heat capacity, thermal conductivity, and tunnelling spectra whilst heat capacity measurements can also be understood with strong-coupling $s$-wave behavior.

In this paper, we report angle resolved DHVA measurements near the upper critical field $B_{c2}$ of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and at low temperatures. Our measurements probe both the "vortex liquid state" and the reduced dimensionality of the electronic properties. We observe DHVA oscillations down to the irreversibility field. The damping of the DHVA signal has been investigated over a wide range of crystal angles up to $\theta = 62^\circ$, and it is found that the $R_s(\theta)$ curves are translated to higher fields as $\theta$ increases, where $\theta$ is the angle between the normal to the conducting layers and the applied magnetic field. The shape of the curves are largely understood in terms of the component of the applied field per-
pendicular to the conducting planes.

II. EXPERIMENTAL DETAILS

The single crystals used in this experiments were grown by a standard electrochemical method. The platelet crystals had well developed facets, making orientation unambiguous. A single crystal was attached with a small amount of vacuum grease onto the end of a flat pick-up coil, which could be rotated in the magnetic field. Two mounting geometries were used: in the first the crystal was rotated in the $a^* - b$ plane; in the second the crystal was rotated by $\phi = 45^\circ$ about the $a^*$ axis and re-mounted on the pick-up coil. Two samples with different size (2 mm and 1 mm in $b$-axis length) from the same batch were examined.

Experiments were performed in a top-loading dilution refrigerator, employing the field modulation technique to measure the DHVA signal, which was detected at the second harmonic of the modulation frequency of 5.2 Hz. Typical modulation amplitudes were in the range $b=10$ mT to 25 mT. AC susceptibility measurements were performed at frequencies of 5.2 Hz and 93 Hz. The external field generated by a solenoid was swept at a rate of 0.001 T/s. The temperature was measured using a calibrated germanium thermometer in the field-compensated region of the mixing chamber.

III. RESULTS

Fig. 1 shows the oscillatory magnetization measured for $T=33$ mK with the magnetic field applied perpendicular to the highly conducting $b$-$c$ planes ($\theta = 0^\circ$). DHVA oscillations are observed above 4 T with a frequency of 601 T (magnified in the inset), originating from the Q2D pockets of the Fermi surface. Fig. 1 shows the in-phase and quadrature components of the AC susceptibility measured at 93 Hz; the inset shows the magnetisation obtained from integrating the in-phase component with field. The feature at 3.9 T in the AC susceptibility corresponds to the irreversibility field, $B_{\text{irr}}$, measured in the torque magnetometry experiments of Sasaki et al.

In this paper, we define the irreversibility field as the field at which there is a rapid increase in the real and imaginary parts of the susceptibility corresponding to the onset of a strong diamagnetic response and pinning of the vortices (see Fig. 2). The relationship of the irreversibility field and the field at which the flux lattice melts remains controversial in this compound.

The DHVA signal is observed only above $B_{\text{irr}}$, in the vortex liquid phase, where the magnetization is fully reversible. The quasiparticle effective mass has been obtained from the temperature dependence of the DHVA signal from the Q2D pockets, and at high fields is found to be $m^* = (3.5 \pm 0.2)m_e$, in agreement with previous studies. We found that the 2D Lifshitz-Kosevich (L-K) expression with constant chemical potential (Eq. 1) describes the field dependence of the amplitude of the DHVA oscillations. The main difference in the functional form of the 2D and 3D L-K expression is a factor of $\sqrt{B}$. In the case of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, the constant chemical potential is provided by the quasi-1D sheets of the Fermi surface, which act as a particle reservoir. The corresponding Dingle plot at 33 mK is shown in Fig. 3. The plot produced a straight line at high fields, from which a Dingle temperature of $T_D = 0.53 \pm 0.02$ K is deduced, which is comparable to other samples. The 2D form of the L-K expression predicts the absolute amplitude of the oscillations, this can be compared with experimental data, by extrapolating to infinite field (1/$B \rightarrow 0$). By assuming the thickness of the 2D layer in Eq. 1 is that of a BEDT-TTF layer, we obtain good agreement with Eq. 1 further validating the use of the 2D L-K expression.

The Dingle plot (Fig. 3) deviates from linearity below $\approx 7$ T, indicating that the DHVA signal suffers an additional attenuation below this field, associated with the onset of superconductivity. In contrast to the behavior in other type II superconductors, this attenuation appears well above the $B_{\text{irr}}$. The additional attenuation can be conveniently expressed as a field-dependent factor amplitude, $R_s(B_0)$, in the L-K formula. The $R_s(B_0)$ values determined from the data in Fig. 3 are shown in Fig. 4(a). $R_s$ curves have been extracted from DHVA data taken at temperatures up to 440 mK, and are shown in Fig. 5. As the temperature is increased, the signal-to-noise is steadily reduced, and this manifests itself as increased data point scatter in the $R_s$ curves. The form of $R_s$ largely unchanged up to $T=440$ mK. The solid curves on the plot will be discussed in the next section.

An angle resolved DHVA study has been performed with the aim of investigating the angular dependence of $R_s$ and the dimensionality of the Fermi surface. Clear DHVA oscillations are observed at high fields, whilst at lower fields a noisy signal due to flux jumps is observed. The DHVA oscillations are observed over a progressively shorter field range as $\theta$ increases, with the position of the lower bound having a $1/\cos \theta$ angular dependence. From the analysis of the DHVA amplitude, both the DHVA frequency and effective mass were found to have a $1/\cos \theta$ angular dependence. These findings are in good agreement with previous studies, demonstrating the 2D nature of the Fermi surface. $R_s$ curves have been extracted at each angle from the deviation of the Dingle plot (Fig. 3) with respect to the straight line. The attenuation due to the superconductivity occurs at higher fields as $\theta$ increases. By considering the components of the main and modulation magnetic fields perpendicular to the $b$-$c$ planes ($B \cos \theta$ and $b \cos \theta$ respectively), the $R_s(\theta)$ curves can be scaled to lie on top of each other, as shown in Fig. 4(a), however, the concave nature is slightly less prominent at higher angles. $R_s(\theta)$ curves showed the same behavior when the crystal was oriented...
such that $\phi = 45^\circ$.

IV. DISCUSSION

A. Additional damping of the DHVA signal in the fluctuation region

The field dependence of the DHVA oscillations in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ differs from other superconductors such as NbSe$_2$ and V$_3$Si in that the damping of the DHVA signal due to the superconductivity appears at a field considerably greater than that where a large diamagnetic response due to the superconductivity is observed. Having said this, if we integrate the AC susceptibility with field to obtain the magnetization, having said this, if we integrate the large diamagnetic response due to the superconductivity and the DHVA signal due to the superconductivity appearing in the introduction, the short coherence length and two-dimensionality of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ mean that fluctuation in the superconducting order parameter will be large over a wide temperature and field range. Thus there is no ‘sharp’ transition into the mixed state at $B_c$.

Rather, $B_{c2}$ should be thought of as the cross-over field associated with the formation of a flux liquid phase. It has been argued that this picture is actually appropriate for all type-II superconductors. The cross over region being smaller in 3D systems.

Understanding the DHVA effect in the superconducting mixed state is a challenging problem from the theoretical viewpoint. The experimental observation of these oscillations has stimulated many theoretical studies over the last ten years. A feature common to many of the theories is the prediction of electronic states within the energy gap, which appear in a plane perpendicular to the applied magnetic field. It is the presence of these states which give rise to the DHVA oscillations in the mixed state. In $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, the presence of a quasiparticle states is supported by thermal conductivity, and heat capacity measurements in a magnetic field. Most of the theories attribute the additional damping of the DHVA signal to the additional scattering of the quasiparticles by the vortex lattice. In the Maki, Stephen approach, the vortex lattice is treated as a random media to scatter the quasiparticles. Dukan and Tesanovic, Norman, MacDonald, and Akera, Manix et al., Zhuravlev et al., Gvozdikov and Gvozdikova, treat the structure of the vortex lattice in detail.

Attempts to model the experimental form of the $R_s$ curves using the available theories describing the DHVA effect in the mixed state, coupled with the mean field form for the field-dependence of the superconducting gap, were unsuccessful in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. The existing theories do not reproduce the downward curvature in $R_s$ observed above about 4.5 T in Fig. 1. A successful explanation of the $R_s$ curves may need to account for the effects of the superconducting fluctuations inherent in this material. One approach is to assume that the DHVA effect is sensitive to the square (or root mean square) of the order parameter $\langle \Delta^2 \rangle$. If the fluctuating part of the order parameter is $\delta \Delta$, then $\langle \Delta^2 \rangle = \langle \Delta \rangle^2 + \langle (\delta \Delta)^2 \rangle$. An approximate form of the fluctuating order parameter can be obtained using the lowest Landau level approximation $\langle \delta \Delta^2 \rangle \propto 1/(B/B_c - 1)$. Far below $B_{c2}$, the superconducting gap takes a mean-field form, $\Delta(B) = \Delta(0) \sqrt{1 - B/B_{c2}}$. We can interpolate between the two limiting regimes with the formula,

$$\langle \Delta^2 \rangle = \sqrt{\left[ \frac{\Delta(0)^2}{2} \left( 1 - \frac{B}{B_{c2}} \right) \right]^2 + \alpha(T)^2}$$

$$+ \frac{\Delta(0)^2}{2} \left( 1 - \frac{B}{B_{c2}} \right).$$

The parameter $\alpha = \langle \Delta^2 \rangle_{B_{c2}}$ determines the strength of the fluctuations at $B_{c2}$, where $\langle \Delta \rangle = 0$. Since the DHVA oscillations are always observed in the vortex liquid state above $B_{irr}$, we employ the Maki, Stephen approach in which the vortices are treated as a random media which scatter the quasiparticles.

$$R_s = \exp \left[ -\frac{\pi^3}{2} \langle \Delta^2 \rangle \left( \frac{m_v e B}{\hbar} \right)^2 \left( \frac{B}{F} \right)^{1/2} \right].$$

The interpolation formula allows the data taken at $T=30$ mK to be fitted with $B_{c2} = 4.8 \pm 0.2$ T and $\alpha = 0.13 \pm 0.05$ meV$^2$ as fitting parameters. In the fitting procedure, the following parameters are used. For the superconducting gap at zero magnetic field, a value $\Delta(0) = 1.5$ meV determined from the BCS relation $2\Delta = 3.5k_BT_c$ with $T_c = 9$ K. For the band mass, a value $m_v = 1.2m_e$ found by cyclotron resonance measurement is used. We obtain a somewhat smaller $B_{c2}$ value compared with $B_{c2} \sim 6T$ that is given by Sasaki et al., based on the magnetic field where torque measurement of the DHVA amplitude start to deviate from the prediction of the normal L-K expression by their torque measurement. In our analysis, we allow the deviation from the normal state expression even above $B_{c2}$ [see Fig. 1(b)], since the effects of fluctuations are taken into account.

Fig. 2 shows $R_s$ curves extracted at temperatures up to $T=440$ mK. The $B_{c2}$ values given by the fluctuation analysis at each temperature are almost unchanged and are in the range of 4.5–5 T, in good agreement with the weak temperature dependence of the $B_{c2}$ in this temperature region.

Fig. 3 summarizes the superconducting phase diagram resulting from the present work. $B_{irr}$ is obtained from the AC susceptibility and the present work. $B_{c2}$ is determined from the DHVA effect via Eq. 1. Because $B_{c2}$ is treated as a “crossover field” defined through
Eq. 2, the values are different from others reported in the literature.

B. The dimensionality of the superconductivity of the $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$

We now discuss the angle dependence of the DHVA signal. The anomaly in the AC susceptibility curve representing the irreversibility field $B_{irr}$ and $B_c$ can be approximately scaled with $B \cos \theta$ as shown in Fig. 8(a). The $B_{irr}$ was found to have a $1/\cos \theta$ angular dependence, in agreement with the angle-dependent torque measurement. This may be taken as evidence for strong 2D nature of the superconductivity in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$.

In Fig. 8(b), the $R_s(\theta)$ curves at each angle of the magnetic field are shown scaled onto one another by taking the component of the magnetic field perpendicular to the layers $B \cos \theta$. The behavior of $R_s$ curves at different angles $\theta$ scales approximately with $B \cos \theta$. Thus the field perpendicular to the conducting layers appears to control the superconducting properties of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. However, in detail, the behavior of $R_s$ curves at different angles $\theta$ is not fully scaled with $B \cos \theta$ near $B_c$. The concave nature of the upper critical field expressed as $B_c^2(\theta) = B_0^2/\cos^2 \theta + \alpha^2$, where $B_0 = (1 + \alpha^2)B_c(\theta = 0)$ and $\alpha = B_0/B_{spin}$. This expression also fits the angle dependence of $R_s$ with $B_{spin} = 30$ T, as shown in Fig. 8(c).

V. CONCLUSION

We have observed the effects of fluctuations and reduced dimensionality on the de Haas-van Alphen (DHVA) signal from $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. The DHVA signal suffers an additional attenuation in the mixed state. The attenuation is analyzed in terms of the superconducting fluctuation effects. Such an analysis can be used to determine the superconducting phase diagram of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. In particular, yielding $B_c$, which we interpret as a cross over field associated with the formation of the vortex flux lattice. The angular dependence of this damping has been measured over a wide range of angles. Our results scale well with the component of applied field perpendicular to the conducting planes suggesting that the superconductivity is highly two dimensional in this material.

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FIG. 1. The oscillatory magnetization as measured by the second harmonic pick-up, with the magnetic field applied perpendicular to the $b$-$c$ planes at a temperature of 33 mK. The DHVA oscillations are observed above 4 Tesla with a DHVA frequency of 601 T. The noisy signal below 3.5 T is due to flux jumps occurring within the sample.

FIG. 2. In-phase $\chi'$ (solid line) and quadrature $\chi''$ (dotted line) components of the AC susceptibility of the sample in a magnetic field applied perpendicular to the $b$-$c$ plane at 32 mK. Inset: The magnetization obtained by integrating the in-phase component of the AC susceptibility. The magnetization is not linear in field above 4 T due to superconducting (diamagnetic) fluctuations.

FIG. 3. Dingle plot of the DHVA signal obtained with the magnetic field applied perpendicular to the $b$-$c$ plane at 30 mK. The deviation in linearity below 7 T represents the additional damping due to superconductivity. $\alpha$ is the amplitude of the DHVA oscillations, $X$ is defined in Eq. 3.

FIG. 4. (a) Damping of the DHVA signal due to superconductivity, $R_s$, at 30 mK in a magnetic field applied perpendicular to the $b$-$c$ plane ($\theta = 0^\circ$). The error bars are caused by the experimental uncertainty in the normal state scattering rate. The solid line is a fit of Eqs. 2–3. (b) The root-mean-square order parameter $\sqrt{\langle \Delta^2 \rangle}$ as given by Eq. 3.
FIG. 5. Fits for the observed $R_s$ to Eqs. 2–3 which include the effects of superconducting fluctuations at $T=30$ mK, together with the data at 130 mK, 240 mK, 350 mK, and 440 mK.

FIG. 6. Superconducting phase diagram for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$: squares are $B_{c2}$ determined by fitting Eqs. 2–3 to DHVA data; triangles are $B_{c2}$ determined from the specific heat anomaly (Ref. 15). The irreversibility field $B_{irr}$ is determined from AC susceptibility measurements: closed circles (this work); open circles (Ref. 13).

FIG. 7. The angular dependence of $B_{c2}$ and $B_{irr}$ determined from DHVA and AC susceptibility measurements respectively. Lines are fits to a $1/\cos \theta$ dependence.
FIG. 8. Scaling of $R_s(\theta)$: (a) assuming $R_s$ depends only on $B \cos \theta$; (b) assuming $R_s$ depends only on $B_{c2}$ with $B_{c2}$ given by Tinkham \cite{Tinkham}; (c) assuming $R_s$ depends only on $B_{c2}$ with $B_{c2}$ given by Nam et al. \cite{Nam}. 