On the absence of spin-splitting in α-(BEDT-TTF)$_2$KHg(SCN)$_4$

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We report the results of a detailed study of the field orientation-dependence of the de Haas-van Alphen waveform in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. By considering the field orientation-dependence of the sign and phase of the fundamental α frequency, at fields both well above and below the kink transition field, it is found that a single value for the product of the effective mass with the electron g-factor can explain the experimental data deep within both the high magnetic field and low magnetic field phases. This implies that spin-splitting does not occur within the low magnetic field phase until the angle between the magnetic field and the normal to the conducting planes is $\sim 42^\circ$. This finding contrasts greatly with that recently published by Sasaki and Fukase, implying that electron-electron interactions do not play a significant role in the formation of the charge-density wave ground state. The manner in which the amplitude of the waveform of the oscillations is damped within the low magnetic field phase is indicative of a non harmonically indexed reduction of the amplitude, thereby eliminating both magnetic breakdown and impurity scattering as dominant damping mechanisms within this phase. Meanwhile, the presence of a large amplitude second harmonic within the low magnetic field phase that has a negative sign over a broad range of angles can only be explained by the frequency doubling effect.

I. INTRODUCTION

Charge-transfer salts of the composition α-(BEDT-TTF)$_2$M$_x$Hg(SCN)$_4$ (where $M = K$, Tl or Rb) exhibit a complex phase diagram that is profusely sensitive both to temperature $T$ and the orientation of an applied magnetic field $B$. The observation of numerous magneto-oscillatory phenomena, as a function of the magnitude and orientation of $B$, has firmly established the existence of a reconstructed Fermi surface below the transition temperature $T_p$, occurring at $\sim 8$ K in the $M = K$ and Tl salts [1] (or $\sim 10$ K in the $M = Rb$ salt). Direct evidence for a lattice superstructure, which would be necessary in order to unambiguously distinguish a charge-density wave (CDW) ground state from one that is a spin-density wave (SDW) [8], however, has not been forthcoming. Only very recently has sufficient indirect evidence been accumulated so as to tip the balance of the argument. For such a simple transition [5], the physical changes incurred experimentally are difficult to reconcile with such a simple transition [8].

Quantum oscillations have proven to be especially sensitive to $B_k$ [13]. All signatures of a reconstructed Fermi surface are lost at high magnetic fields [2,14,20], while the effective mass $m^*$ of the dominant $\alpha$ frequency appears to increase [14,15]. Most notable, perhaps, is the change in the physical appearance of the waveform from one that is strongly damped but displaying split maxima at fields below $B_k$ [1,2,12], to one that is triangular at fields above $B_k$ [1,2,24]. The origin of the splitting of the waveform within the CDW$_0$ phase is, itself, a contemporary issue. Numerous publications [1,2,22] attest to the fact that this splitting appears to resemble the “spin-splitting” phenomenon that occurs when the degree to which the Landau levels are split by the Zeeman energy, $\Delta \varepsilon = gheB/m_e$ ($g$ being the electron $g$-factor and $m_e$ being the free electron mass), becomes commensurate with an odd half-integer multiple of the cyclotron energy, $\hbar \omega_c = \frac{eB}{m^*}$ [27]. The observation of a split waveform does not, however, prove the existence of spin-splitting. A similar split waveform occurs in the CDW compound NbSe$_3$ [24], for example, yet this was recently shown not at all to be related to the Zeeman effect [27]. The frequency doubling (FD) effect [28], which occurs when an additional term proportional to the square of
the oscillatory chemical potential $\tilde{\mu}$ modulates the free energy of a CDW ground state, could provide an alternative explanation. In spite of the fact that seemingly strong arguments have been given for spin-splitting in the $\alpha$-(BEDT-TTF)$_2$Mg(SCN)$_4$ salts [1,2,22,29], this hypothesis has not actually been thoroughly tested.

The purpose of the present paper is to thoroughly investigate the possibility of both the spin-splitting and FD effects in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ by performing a detailed investigation of the field orientation-dependence of the waveform at a variety of magnetic fields both above and below $B_k$. In this work, the product $\nu_0^* g$ of the degree to which the effective mass is enhanced $\nu_0^* = m_e^*/m_0$ with the electron $g$-factor is determined by fitting to the field orientation of the fundamental oscillation sign and phase. As will be discussed in Section III, this is required in order to determine the relative importance of electron-electron ($e-e$) and electron-phonon ($e-ph$) interactions in the formation of the ground state. The validity of the canonical ensemble for describing the field-orientation dependence of the waveform within the high magnetic field phase is discussed in Section IV, while the details concerning the anomalous behaviour of the quantum oscillations within the low magnetic field phase are described in Section V. We turn to a discussion of the frequency doubling effect in Section VI and summarise the results in Section VII.

II. EXPERIMENTAL

The single crystal sample of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ of volume $\sim 0.8 \text{ mm}^3$, used in this study, was the same as that used for the magnetic torque measurements in Reference [1]. It was mounted on the moving plate of a phosphor bronze capacitance cantilever, which was itself attached to a rotating platform for which the axes of torque and rotation were parallel to each other, yet both perpendicular to $\mathbf{B}$. The angle between $\mathbf{B}$ and the normal to the capacitance plates was approximately the same as the angle $\theta$ between $\mathbf{B}$ and the normal $\mathbf{n}$ to the conducting planes of the sample. The capacitance, of order $\sim 1 \mu$F, was measured by means of a capacitance bridge energized with 30 V rms at 5 KHz and was observed to change by less than 0.1 %. Since this implies a maximum angular displacement of $\sim 1.5^\circ$, torque interaction effects were insignificant. Static magnetic fields extending to $\sim 32 \text{ T}$ were provided by the National High Magnetic Field Laboratory, Tallahassee, while a constant temperature of $\sim 450 \text{ mK}$ was obtained using a $^3\text{He}$ refrigerator.

Because the interlayer transfer integral $t_\perp$ of $\alpha$-(BEDT-TTF)$_2$Mg(SCN)$_4$ charge-transfer salts is immeasurably small compared to those within the planes, these materials provide some of the best known examples of ideal multilayered two-dimensional (2D) Fermi liquids [23]. The only significant component of the Landau diamagnetic susceptibility is that projected along $\mathbf{n}$. Because $\tau = \mathbf{M} \times \mathbf{B}$, the oscillatory component of the magnetic torque is

$$\tilde{\tau}_\theta = -\tilde{M}_{\perp,\theta} B \sin \theta,$$

where $\tilde{M}_{\perp,\theta}$ is the oscillatory component of $\mathbf{M}$ parallel to $\mathbf{n}$.

III. FIELD ORIENTATION-DEPENDENCE OF THE DHV A PHASE

Examples of the oscillatory magnetic torque of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$, measured in static magnetic fields of up to 32 T and at several different field orientations, are shown in Fig. 1. Note that these data closely resemble earlier measurements made on the same material [18,19]. A Fourier transformation of the data over a restricted range of $B$ (in the $1/B$ domain) within the low magnetic field phase at $\theta = 8.8^\circ$, shown in Fig. 2, reveals a plethora of harmonics indicative of a good sample quality.

A number of recent articles have shown that the oscillations of $\tilde{\mu}$ (in this and other 2D materials) invalidates a simplistic analysis of the dHvA oscillation data in terms of the Lifshitz-Kosevich (LK) formula [23]. The reasons for this are twofold. First, the LK formula is suited only to systems in which the Fermi surface is significantly curved in all three $k$-spatial dimensions [23,25]. Second, the oscillations of the chemical potential $\tilde{\mu}$ significantly perturb the waveform of the oscillations so as to cause the amplitude and sign of each of the $p > 1$ harmonics to depart significantly from those predicted by the LK model [23]. Further complications may also arise as a result of interactions involving the condensate itself, due either to magnetic breakdown [24], the FD effect [25] or induced currents. Induced currents, that contribute an additional oscillatory structure to the dHvA waveform, have now been shown to occur both in static magnetic fields [26] and pulsed magnetic fields [27,28].

In spite of the fact that the waveform of the dHvA oscillations is significantly perturbed by $\tilde{\mu}$ in the canonical ensemble, the underlying sign and phase of the fundamental frequency (which we shall label as $p = 1$) is the same as that in the grand canonical ensemble (or equivalently the LK model) for which $\tilde{\mu}$ is assumed to be 0 [23,25]. The amplitude of the fundamental oscillations in the magnetic torque can therefore be written in the form

$$\tilde{\tau}_{1,\theta} \approx A_{1,B,T,\theta} \sin \left( \frac{2\pi F}{B} \right) S_{1,\theta} \sin \theta,$$

where $A_{1,B,T,\theta}$ is a monotonically varying sign and phase independent prefactor (for which there is no simple algebraic form in the canonical ensemble) and $F$ is the dHvA
frequency. Only the Zeeman term $S_{1,\theta} = \cos(\pi \nu_0^* g / 2)$ determines the sign and phase of the oscillations [24], for which $\nu_0^* = m_e^* / m_e = m_0^* / m_e \cos \theta$ [24].

The magnitude $|S_{1,\theta}|$ becomes unity whenever $\Delta_e$ becomes commensurate with $\hbar \omega_c$, or, equivalently, when the product $\nu_0^* g$ becomes equal to an even integer. Conversely, whenever $\nu_0^* g$ is equal to an odd integer, the amplitude of the oscillations depends on $\nu_0^* g / \cos \theta$, becoming commensurate with $\hbar \omega_c$. Until a node becomes commensurate with $\hbar \omega_c$, the positions of the nodes are not exactly reproduced.

By the dashed line, the positions of the nodes are not accurately reproduced. However, in disagreement with the results of Sasaki and Fukase [29], their reported value of $\nu_0^* g = 4.7$ within the CDW phase requires the existence of a node at $\theta = 20^\circ$ that has never actually been observed. The data in Fig. 3b shows no evidence for either an abrupt change in $\nu_0^* g$ or the development of a node at $\theta = 20^\circ$. Sasaki and Fukase did not attempt to verify the presence or absence of a node at this orientation [29].

A reliable extraction of $\nu_0^* g$ is required for determining the relative importance of $e-e$ and $e-ph$ interactions. According to Fermi liquid theory, $e-e$ and $e-ph$ interactions affect $\nu_0^* g$ and $g$ differently [28]. In the case of $e-e$ interactions, an increase in $\nu_0^* g$ is approximately offset by a reduction in $g$ so that there is no overall change in the product $\nu_0^* g$. This is not, however, the case with $e-ph$ interactions. We can therefore conclude that, since $\nu_0^* g$ does not change appreciably in this experiment, there is no significant change in $e-e$ interactions on passing between the CDW phases, thereby contrasting with the conclusion reached by Sasaki and Fukase [29].

One important question that remains to be answered, therefore, is whether the apparent change in the effective mass of the $\alpha$ frequency on crossing $B_k$ is genuinely related to a change in the strength of the $e-ph$ interactions [24, 19]. Under normal circumstances, dHvA data is more reliable owing to the fact it is derived from a thermodynamic function of state. However, there also exists the possibility that gaps of order $2\Delta$ in the energy spectrum resulting from the formation of the CDW state, lead to breaks in the $\alpha$ orbit trajectory that then have to be overcome by magnetic breakdown in a magnetic field [19]. It has been argued that since $2\Delta$ falls with increasing temperature, this should lead to an additional temperature-dependent term in the quantum oscillation amplitude that could potentially cause the effective mass within the CDW phase to appear artificially low [23]. In Section V, however, we will show that magnetic breakdown appears not to be the dominant form of damping within the CDW phase.
phase. In any case, the reported difference in the degree to which the effective mass is enhanced between the CDW$_0$ and CDW$_x$ phases, $\delta \nu_0^x \sim 0.5$, appears to be quite significant. Within the CDW$_0$ phase, dHvA measurements all agree that $\nu_0^x \sim 1.5 m_e$. Within the CDW$_x$ phase, however, only one estimation of $\nu_0^x \sim 2.0 m_e$ has been made that properly accounts for the effects of induced currents, now shown to occur both in static and pulsed magnetic fields [5]. Since $\alpha$-(BEDT-TTF)$_2$KHz(SCN)$_4$ is believed to possess a CDW ground state, of some form [1 2 3 29], changes in $\nu_0^x$ between CDW sub-phases could be expected. A common observation in all CDW materials is that gaps open in the phononic density of states as well as in the electronic density of state, often referred to as the Kohn anomaly [8]. Since the net e-ph coupling strength is determined by an integration over both the phononic and electronic densities of states [23], an increase in the effective mass should be expected on passing into a phase within which 2$\Delta$ is lower.

This still leaves the behaviour within the narrow field region, 18.2 $B$ < 20.3 T, unexplained. One likely possibility is that the analysis procedure is not able to separate contributions to the dHvA effect originating from the different CDW$_0$ and CDW$_x$ phases over this range, owing to the possible existence of one or more first order changes in the electronic structure just below $B_k$ [23]. We also note that at higher angles, $|\theta| \gtrsim 45^\circ$, another phase, CDW$_y$, has been proposed to exist [4], which could only complicate matters further.

IV. DHVÀ WAVEFORM WITHIN THE CDW$_x$ PHASE

That the conventional form of $S_{1,\theta}$ is well obeyed both in the CDW$_0$ and CDW$_x$ regimes of $\alpha$-(BEDT-TTF)$_2$KHz(SCN)$_4$, (with the exception of a narrow field interval immediately below $B_k$), implies that this material has, at all times, a well defined set of Landau levels characteristic of a normal Fermi liquid in a magnetic field. Given, also, that the product $\nu_0^x g$ is close to an integral value (i.e. 4), it is of no surprise that the high magnetic field phase closely resembles a canonical ensemble of electrons for which the spins are approximately degenerate [23]. On inserting more exact parameters, $\alpha^* \sim 2.0$ [2] and $\nu_0^x g \sim 3.67$ (this work) into the numerical model of Reference [23], the canonical ensemble (calculated in Fig. 4b) is able to reproduce the experimentally observed magnetic torque in Fig. 4a rather well. The data in Fig. 4a was taken in a dilution fridge at $T \sim 27$ mK and $\theta \sim 7^\circ$. Note that the parameters $F$, $\nu_0^x$, $\gamma$ and $\nu_0^x g$ are constants specific to the material that have been determined experimentally and cannot be arbitrarily adjusted as fitting parameters. Only the scattering rate $\tau^{-1} \sim 0.6 \times 10^{12}$ s$^{-1}$, which is always sample-dependent, can be adjusted in order to obtain the best representation of the experimental data.

These same parameters, when inserted into the numerical canonical ensemble calculation, are also able to reproduce the correct field orientation-dependence of the fundamental $(p = 1)$ amplitude of the magnetic torque in Fig. 4a, at least for $|\theta| \lesssim 45^\circ$. The same numerical model also predicts the correct sign of the second $(p = 2)$ harmonic, albeit that the field orientation-dependence of its amplitude is less accurately reproduced. By “sign” we refer to the sign of $a_p$, that correctly depicts the waveform of the oscillations when it is decomposed into a Fourier expansion of the form

$$\tilde{M} \approx \sum_p a_p N \beta^* \sin \left( \frac{2\pi p F}{B} \right).$$

Here, $\beta^* = \hbar e/m^* = \hbar e/m^*$ is the double Bohr magneton, $N$ is the density of carriers giving rise to the $\alpha$ frequency quantum oscillations and $|a_p| < 1$ represents the degree to which the amplitude of each harmonic is attenuated due to the combined effect of impurities, spin and temperature. In the canonical ensemble, there is no simple way to separate each of these contributions [23]. The field orientation-dependence of the sign of the second harmonic is expected to remain positive in the canonical ensemble for all angles when $\gamma > 0.5$ [23] (i.e. when the density of states of the 2D Fermi surface pocket is larger than that of the quasi-one-dimensional sheets). In Fig. 4a we can see that, while the grand canonical ensemble (i.e. the LK model which assumes a fixed chemical potential) is equally well able to explain the behaviour of the fundamental, it fails to account for the positive sign of the second harmonic. This illustrates the hazards associated with fitting the LK model to a 2D system for which it does not apply [17–19,33]. In Section V we will show that this issue becomes particularly important when attempting to understand the oscillations within the CDW$_0$ phase.

In spite of the fact that the models are able to predict the correct form of the fundamental amplitude of the dHvA oscillations at small angles, they cannot account for their rapid attenuation at larger angles, $|\theta| \gtrsim 45^\circ$, in Fig. 4a. One possible explanation is that the scattering rate $\tau^{-1}(k)$ is strongly dependent on $k$, with “hot spots,” or possibly even “hot bands,” occurring at certain values of $k_z$ (the lattice vector parallel to $\mathbf{n}$) [37]. Such effects have been suggested to be important in some of the Bechgaard salts [38]. Since a dHvA experiment senses only a weighted average of $\tau^{-1}(k)$, the number of orbits that intersect hot regions of the Fermi surface could increase for large $\theta$. It was noted in Reference [39], that the experimentally observed scattering rate appears to increase roughly in proportion to tan $\theta$. This damping of the oscillations at large angles will be the subject of a future publication [37].
V. DHVA WAVEFORM WITHIN THE CDW<sub>0</sub> PHASE

Thus far, we have shown that at least two parameters associated with the α frequency appear not to change on traversing B<sub>0</sub>; namely, its fundamental frequency \( F \sim 670 \text{ T} \) and the product \( \nu_0 g_\alpha \). The same cannot be said with confidence about the degree to which the effective mass is enhanced \( M_0 \), the \( \gamma \) parameter (which quantifies the fraction of the density of states occupied by the 2D pocket), or the scattering rate \( \tau^{-1} \). A number of groups have reported an apparent increase in the scattering rate within the low magnetic field CDW<sub>0</sub> phase with respect to that within the high magnetic field phase \([7,8]\). Others have attributed the loss of amplitude of the α frequency within the CDW<sub>0</sub> phase to magnetic breakdown effects \([9,10]\). While the latter might be expected following the introduction of an additional periodic potential \( 2\Delta_0 \) within the CDW<sub>0</sub> phase \([11,12]\), neither of these two possibilities can satisfactorily explain the experimental data. For either of them to be true, the field dependence of the amplitude of each harmonic \( p \) (having corrected for its temperature dependence) would have to be proportional to \( R_{p,B} \approx \exp(-p\bar{\Upsilon}/B) \), where \( \bar{\Upsilon} \) represents the total degree of damping inclusive of both effects. In Fig. 3, the experimentally observed fundamental amplitude of the oscillations within the low magnetic field phase, at \( B \sim 16.5 \text{ T} \), can be approximately reproduced using the numerical model by setting \( \bar{\Upsilon} \sim 79 \text{ T} \). This is equivalent to a scattering rate of \( \tau^{-1} \sim 2.9 \times 10^{12} \text{ s}^{-1} \), comparable to that obtained in Reference \([13]\), or, alternatively, to a characteristic magnetic breakdown field of \( B_0 \sim 79 \text{ T} \). Implicit to either of these explanations, however, is the reduction of the amplitude of the second \( (p = 2) \) harmonic with respect to that of the fundamental by another factor of approximately \( R_B \approx \exp(-\bar{\Upsilon}/B) \sim 10^{-2} \). This appears not the case experimentally, however. For example, when we calculate the waveform using the numerical model (with \( \bar{\Upsilon} \sim 79 \text{ T} \)), the amplitude of the second harmonic in Fig. 3 is roughly two orders of magnitude smaller than that detected experimentally. This would also be the case were we to calculate the waveform using the grand canonical ensemble, or were we to take into consideration FD effects (see Section VI). Clearly, the presence of a second harmonic with an amplitude that is measured to be an appreciable fraction of that of the fundamental is inconsistent with a harmonically indexed damping factor of the form \( R_{p,B} \approx \exp(-p\bar{\Upsilon}/B) \) with \( \bar{\Upsilon} \) being as large as 79 T. We can therefore eliminate both impurity scattering and magnetic breakdown as dominant mechanisms for the damping of the the dHvA oscillations observed within the CDW<sub>0</sub> phase, since both of these lead to harmonically indexed damping factors. The only alternative explanation, therefore, is that the quantum oscillations within the CDW<sub>0</sub> phase is uniformly suppressed in amplitude in a manner that is not indexed to the harmonics. Evidence for a non harmonically indexed reduction of the amplitude has already been published \([7]\). In Reference \([13]\), the Dingle plots of the fundamental and second harmonic were found to have approximately the same slope, indicating the existence of a amplitude reduction factor that is not indexed to \( p \).

In order to account for these experimental observations, we can notionally introduce a damping factor of the form \( R'_B \approx \exp(-\Upsilon'/B) \) within the CDW<sub>0</sub> phase that is independent of \( p \) but that operates in addition to the conventional damping that occurs within the high magnetic field phase. An exponential form for \( R'_B \) is required in order to account for the fact that the Dingle plots are linear \([7]\).

The most trivial interpretation of a non harmonically indexed damping factor is that where the effective volume of the sample contributing to the dHvA signal is reduced by a factor \( R'_B \approx \exp(-\Upsilon'/B) \). A volume reduction factor of this type could, for example, be expected were the CDW<sub>0</sub> phase composed of two coexisting phases spatially separated over distances larger than the cyclotron length, only one phase of which yields dHvA oscillations of the α frequency, with their composition then changing with field. When the dHvA waveform in Fig. 3 is calculated using the same material parameters as within the CDW<sub>x</sub> phase, but with an additional empirical damping term of the form \( R''_B \approx \exp(-\Upsilon''/B) \), setting \( \Upsilon'' \sim 60 \text{ T} \), the model is able to reproduce the experimentally observed amplitudes somewhat better than in Fig. 3. In particular, the model now predicts the second harmonic to have the correct order of magnitude, albeit that is has the opposite sign to that observed experimentally. We will return to a discussion of the sign of the second harmonic in Section VI where we consider FD effects.

Above we have shown that neither impurity scattering nor magnetic breakdown can account for the strong damping within the CDW<sub>0</sub> phase. To carry this argument further, it can be shown that both of these explanations are unphysical for other reasons. For example, a scattering rate is usually determined by the number of defects and impurities in a metal, and this number is not expected to change across a phase transition. The product \( m^* \tau^{-1} \) invariably remains constant. Similarly, the estimated value of \( \Upsilon \sim 79 \text{ T} \) significantly exceeds the magnetic breakdown field \( B_0 \approx n\hbar^2 g_\text{m} \beta / 2e\mu_0 c \sim 20 \text{ T} \) that should be expected for \( n \sim 6 \) magnetic breakdown nodes of size \( \varepsilon_{\text{gap}} \sim 2\Delta_0 \approx 4 \text{ meV} \); \( \varepsilon_F = \hbar eF/m^* \).
being the Fermi energy.

VI. FREQUENCY DOUBLING

The most distinguishing feature of the oscillations in the magnetic torque within CDW$_0$ phase is the presence of a strong second harmonic. The ratio of the harmonics is unaffected by the uniform non harmonically indexed reduction in the amplitude of the oscillations discussed in the preceding section. Another important feature of the dHvA oscillations within the CDW$_0$ phase, which has not been addressed by earlier publications, is that the sign of the second harmonic is negative compared to one that is positive above $B_k$. The change in sign of the second harmonic between the low and high magnetic field phases gives rise to a node at $B_k$ as observed by Uji et al. [19].

The negative sign of the second harmonic within the CDW$_0$ phase is clearly unexpected in the canonical ensemble. It is also inconsistent with spin-splitting in the grand canonical ensemble (or LK model), for which a field orientation dependence of the waveform within the CDW$_0$ phase is expected to be negative when frequency doubling effects are taken into consideration.

In order to model the extent to which the FD effect can affect the waveform, it is useful to consider the proportionality $\tilde{\mu} = BM/N$ [23,10] which, when combined with Equation (1), enables the oscillations in the chemical potential to be written as a series expansion of the form

$$\tilde{\mu} \approx \sum_{p} \frac{a_p \hbar \nu_p}{\pi p} \sin \left( \frac{2\pi p F}{B} \right).$$

According to the frequency doubling model, oscillations in the chemical potential give rise to an additional term in the free energy of the form $\tilde{\Phi}_{FD} = g_{1D} \tilde{\mu}^2$ where $g_{1D}$ is the density of quasi-one-dimensional states that become nested [28]. If we assume the limit $a_1 \gg a_2$ and count only oscillatory terms, this free energy can be written as

$$\tilde{\Phi}_{FD} \approx -\frac{g_{1D} (a_1 \hbar \nu_p)^2}{2\pi^2} \cos \left( \frac{4\pi p F}{B} \right).$$

The resulting frequency doubling term in the magnetisation, $M_{FD} = -\partial \tilde{\Phi}_{FD}/\partial B$, thus has the form

$$M_{FD} \approx -\frac{2a_1^2 N \beta^* g_{1D}}{\pi g_{2D}} \sin \left( \frac{4\pi p F}{B} \right),$$

where $g_{2D} = N \beta^*/F$ is the total density of 2D states. Note that the sign of $M_{FD}$ is negative and can have an amplitude as much as four times larger than the amplitude of the conventional dHvA contribution to the second harmonic. On inserting $g_{1D}/g_{2D} \approx 1$ into the expression

$$\frac{M_{FD}}{M_1} \approx -2a_1 \frac{g_{1D}}{g_{2D}},$$

for the harmonic ratio, the correct amplitude and sign of the second harmonic can be approximately reproduced in Fig. 6 over a wide range of angles, with the exception of the interval $15^\circ < \theta < 40^\circ$. Only the FD model can account for the negative sign of the second harmonic over a wide range of orientations. While there exists some departure from the predictions of the FD model in the range $15^\circ < \theta < 40^\circ$, this is likely to be related to a similar anomaly in the amplitude of the fundamental over the same angular range.

VII. CONCLUSION

In summary, we have shown that a single value of $\nu_0^g$ can account for the field-orientation of the sign and phase of the dHvA oscillations deep within both the CDW$_0$ and CDW$_x$ phases above and below $B_k$. The implications of this are twofold: first, the split waveform that occurs within the CDW$_0$ phase for field orientations $\theta < 42^\circ$ cannot be attributed to spin-splitting, and second, the role of $e-e$ interactions appears not to change between the two phases.

We have also shown that the field orientation dependence of the waveform within the CDW$_x$ phase is entirely consistent with the predictions for a canonical ensemble of electrons with a background reservoir of quasi-one-dimensional states. The behaviour of the waveform within the CDW$_0$ phase, however, is more unusual. The waveform of the oscillations appears to be significantly reduced in amplitude uniformly across the harmonics. Since this damping is not indexed to the harmonics, we can eliminate both impurity scattering and magnetic breakdown as dominant mechanisms for the reduction of the amplitude within the CDW$_0$ phase. Rather, it appears to be the case that the effective volume of the sample contributing to the dHvA signal is field dependent within the CDW$_0$ phase.

It is shown that the negative sign of the second harmonic that occurs within the CDW$_0$ phase over a large range of angles cannot be explained in terms of the dHvA effect. A negative sign is expected to follow naturally from the frequency doubling effect, however. The presence of frequency doubled oscillations in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ is consistent with the existence of a commensurate CDW ground state [3].
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FIG. 1. Examples of the oscillatory magnetic torque at several different angles measured in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at 450 ± 20 mK throughout. The traces have been offset with respect to each other for clarity.

FIG. 2. Fourier transform of the data at $\theta \sim 8.8^\circ$ in Fig. 1 over a restricted range of field ($18.2 < B < 23$ T).

FIG. 3. (a) Field orientation-dependence of the Fourier amplitudes at different magnetic fields in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. Bezier fits between the points are shown for clarity. (b) The field orientation-dependence of the quantum oscillations at $B \sim 26.5$ T together with the functional form of $S_{1,0} \sin \theta$ best able to reproduce the correct positions of the nodes drawn as a solid line. (c) The field orientation-dependence of the quantum oscillations at $B \sim 16.5$ T, with the function form for $S_{1,0} \sin \theta$ shown with $\mu^* g \sim 3.67$ (solid line) and 4.7 (dotted line) respectively.

FIG. 4. (a) An example of the oscillations in the magnetic torque measured in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at low temperatures ($T \sim 27$ mK) having subtracted the induced currents as described in Reference [5]. (b) The calculated waveform of the oscillations, using the canonical ensemble as described in the text.

FIG. 5. (a) Field orientation-dependence of the amplitude of the fundamental $p = 1$ and second harmonic $p = 2$ in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at 26.5 T, together with those calculated using to the canonical ensemble. (b) The same data but with the fundamental and second harmonic calculated using the grand canonical ensemble (i.e. the LK model).

FIG. 6. (a) Field orientation-dependence of the amplitude of the fundamental $p = 1$ and second harmonic $p = 2$ in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ at 16.5 T, together with those calculated using the canonical ensemble with $\Upsilon = 79$ T, which equates to an effective scattering rate of $2.9 \times 10^{12}$ s$^{-1}$. (b) The same data but with the fundamental and second harmonic calculated using the same scattering rate as within the CDW$_x$ phase, but with $\Upsilon' = 60$ T.
\[ \theta = 8.8^\circ \]
\[ \theta = 28.8^\circ \]
\[ \theta = 48.8^\circ \]
\[ \theta = 68.8^\circ \]

Figure 1 of Harrison et al.
Figure 2 of Harrison et al.

\[ \theta = 8.8^\circ \]

\[ 18.2 < B < 23 \, \text{T} \]
Figure 3a of Harrison et al.

- $\tau$ (Arb. Units)
- $\theta$ ($^\circ$)

- 26.5 T
- 23.0 T
- 20.3 T
- 18.2 T
- 16.5 T
- 15.0 T
Figure 3b of Harrison et al.
$B = 16.5 \text{ T}$

Figure 3c of Harrison et al
Figure 4 of Harrison et al.

(a) Experimental data ($T = 27 \text{ mK}$)

(b) Canonical ensemble calculation
Figure 5 of Harrison et al.

(a) canonical ensemble

(b) grand canonical ensemble
Figure 6 of Harrison et al

(a) Canonical ensemble

\[ \Upsilon = 79 \text{ T} \]

(b) Canonical ensemble

\[ \Upsilon' = 60 \text{ T} \]