Anomalous Quantum Oscillations in a Heterostructure of Graphene on a Proximate Quantum Spin Liquid

V. Leeb,1 K. Polyudov,2 S. Mashhadi,2 S. Biswas,3 Roser Valentí,3 M. Burghard,2 and J. Knolle1,4,5

1Department of Physics TQM, Technische Universität München, James-Franck-Straße 1, D-85748 Garching, Germany
2Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany
3Institut für Theoretische Physik, Goethe-Universität Frankfurt, 60438 Frankfurt am Main, Germany
4Munich Center for Quantum Science and Technology (MCQST), 80799 Munich, Germany
5Blackett Laboratory, Imperial College London, London SW7 2AZ, United Kingdom

(Dated: October 6, 2020)

The quasi two-dimensional Mott insulator α−RuCl3 is proximate to the sought-after Kitaev quantum spin liquid (QSL). In a layer of α−RuCl3 on graphene the dominant Kitaev exchange is further enhanced by strain. Recently, quantum oscillation (QO) measurements of such α−RuCl3 / graphene heterostructures showed an anomalous temperature dependence beyond the standard Lifshitz-Kosevich description. Here, we develop a theory of anomalous QO in an effective Kitaev-Kondo lattice model in which the itinerant electrons of the graphene layer interact with the correlated magnetic layer via spin interactions. At low temperatures a heavy Fermi liquid emerges such that the neutral Majorana fermion excitations of the Kitaev QSL acquire charge by hybridising with the graphene Dirac band. Using ab-initio calculations to determine the parameters of our low energy model we provide a microscopic theory of anomalous QOs with a non-LK temperature dependence consistent with our measurements. We show how remnants of fractionalized spin excitations can give rise to characteristic signatures in QO experiments.

Introduction. Quantum oscillation measurements are a standard tool for determining the electronic structure of metallic materials. Famously, Onsager showed how the oscillation frequency of the magnetization or resistivity as a function of inverse magnetic field are directly related to a metal’s Fermi surface [1]. The standard QO theory was then completed in 1956 by Lifshitz and Kosevich who derived their well-known LK-formula [2] for the temperature dependence of the amplitude decay which permits an extraction of the effective mass. Over the following decades, one by one all elementary metals followed this canonical description [3]. Even strongly correlated systems like heavy fermion metals [4] and cuprate high temperature superconductors were no exceptions, i.e. their QO amplitudes showed LK-behaviour and the main sign of correlation effects are effective mass enhancements [5].

The observation of QOs with a non-LK temperature dependence in the correlated insulator SmB6 [6, 7] came totally unexpected, challenging the canonical description of QO. It initiated the search for QO in other correlated insulators, i.e. YbB12 [8, 9], and motivated theoretical works [10, 11] which unearthed scenarios beyond the LK paradigm. For example, inverted band insulators could lead to such anomalous QO with a non-LK temperature evolution [10], which were then predicted [12] and subsequently observed in quantum well heterostructures [18, 19]. Alternatively, it was shown that charge neutral fermions from fractionalization in strongly correlated insulators can potentially give rise to QOs via indirectly coupling to the orbital magnetic field [14, 15]. The latter idea was originally introduced for QSLs [20], which are quantum disordered long-range entangled magnetic phases [21, 22]. While the debate about SmB6 is not settled, an exciting development is the advent of heterostructures with two-dimensional magnets [23], which pave the way for studying novel QO phenomena from the interplay of magnetic fluctuations and itinerant charges.

Heterostructures of α−RuCl3 on graphene have been recently synthesized [24, 25], see Fig.1(a) and (b), which caused considerable excitement because the insulating α−RuCl3 layer is believed to be in proximity to a QSL described by the seminal Kitaev honeycomb model [26–30]. The heterostructure undergoes hole (electron) doping of the graphene (Ru) layer [24, 25] due to a charge transfer between the correlated insulating layer and itinerant graphene [21, 32]. The lattice-mismatch-induced strain is expected to increase the relevance of the Kitaev spin exchange in the magnetic layer [31, 33] bringing the system closer to the Kitaev QSL. Intriguingly, resistivity measurements as a function of magnetic field show anomalous QO with a non-LK temperature dependence. Instead of a monotonic decay as a function of increasing temperature, a maximum appears at a temperature of around 7 K which has been suggested to originate from the magnetic fluctuations of the Kitaev magnet [25].

Here, we develop a microscopic theory of anomalous QO in a Kitaev–graphene heterostructure. We provide new QO measurements on graphene in proximity to α−RuCl3 and show that our quantitative theory is consistent with their distinct non-LK behaviour. Thus, we establish that the interplay of fractionalized spin excitations and itinerant electrons can lead to anomalous QO beyond the LK paradigm.
Our strategy is as follows: We first construct a minimal two-layer model of a Kitaev system coupled to a graphene lattice via spin interactions. Previous works \[24, 33\] showed within a Majorana mean-field theory (MFT) that such a Kitaev-Kondo lattice hosts strongly correlated phases like fractionalized Fermi liquids or p-wave superconductivity. In addition, a heavy Fermi-liquid (hFL) phase can be stabilised in which the Majorana fermions of the Kitaev QSL effectively hybridise with the itinerant electrons from the graphene layer. To connect to our measurements, we extract the microscopic parameters of the hFL phase from a tight-binding fit to the ab-initio band structure of Ref. \[33\], see Fig 1(c). From a low energy expansion of our model we then calculate the exact form of the Landau level (LL) structure, which is used to derive an analytic formula for quantum oscillations. As the main result, we show that the oscillation frequency and distinct temperature dependence of our theory can explain the anomalous QO behaviour of the $\alpha$–RuCl$_3$ / graphene heterostructure.

**Effective Low Energy Model.** Our starting point is a monolayer Kitaev honeycomb spin model on top of a graphene honeycomb layer (lattice vectors $n_1, n_2$ and lattice constant $a$). For simplicity we assume that both layers are commensurate but we comment on the effect of lattice mismatch below. The key ingredient is the form of the interaction between the two layers. Because of the Mott insulating nature of $\alpha$–RuCl$_3$ we model it as a spin-only Kondo coupling such that the Hamiltonian $H = H_K + H_t + H_J$ is a sum of three terms:

$$H = -K \sum_{(ij)\alpha} S_i^\alpha \alpha S_j^\alpha - t \sum_{(ij)\sigma} \langle c_i^{\dagger \alpha} c_j^{\alpha \sigma} + h.c. \rangle + J \sum_{i,\alpha} c_i^{\dagger \alpha} \tau^{\alpha \sigma} c_i^{\alpha \sigma} S_i^\alpha,$$

(1)

$S^\alpha$ are the spin-$\frac{1}{2}$ operators (components $\alpha = x, y, z$ also label the inequivalent bond directions $(ij)\alpha$ on the honeycomb lattice). The $c_i^{\dagger \alpha}$ create conduction electrons with spin $\sigma$ and $\tau^{\alpha}$ are the Pauli matrices.

The first term is the usual Kitaev model with competing Ising exchanges of isotropic strength $K$ along the inequivalent bond types. The exact solution of $H_K$ proceeds by decomposing the spin operators into real Majorana fermions $\chi^\alpha_i, S_i^\alpha = i\chi^0_i \chi^\alpha_i$ \[23\]. An important observation for our purpose is that the ground state flux sector can be described exactly with a Majorana MFT \[32–34\]. The second term of the Hamiltonian describes the nearest-neighbour hopping with strength $t$ of the electrons. The last part is the Kondo coupling with parameter $J$ between localized spins and itinerant electrons.

To treat the Kitaev-Kondo lattice we follow previous work \[34\] and rewrite the complex fermions $c_{i,\alpha} = \frac{1}{\sqrt{2}} (\eta_{i,\alpha}^0 + i\eta_{i,\alpha}^1)$ and $c_{i,\downarrow} = \frac{1}{\sqrt{2}} (i\eta_{i,\downarrow}^1 - \eta_{i,\downarrow}^0)$ into a sum of Majorana fermions. Then we introduce real mean-fields $U^\mu = (i\chi^\mu_i \chi^\nu_j)$ and $W^\mu = (i\eta^\mu_i \eta^\nu_j)$ such that the MFT Hamiltonian reads (dropping all constants),

$$H = \sum_{i,\alpha} \sum_{\lambda} \left( c_{k,\lambda,\alpha} \right)^\dagger W^{\mu_1 \nu_1} t_{\theta_k} W^{\mu_2 \nu_2} \left( c_{k,\lambda,\alpha} \right) + \sum_{ij} \sum_{\sigma,\tau} \left( c_{k,\lambda,\alpha} \right)^\dagger W^{\mu_1 \nu_1} t_{\theta_k} W^{\mu_2 \nu_2} \left( c_{k,\lambda,\alpha} \right)$$

(2)

where the spin matrices $M^{\alpha}$ are given by $M^1 = \tau^1 \otimes i\tau^2, M^2 = \tau^2 \otimes \tau^0$ and $M^3 = \tau^1 \otimes \tau^2$.

In the hFL phase the mean-fields are reduced to $U = u \mathbb{1}$ and $W = w \mathbb{1}$ \[38\] which allows us to write the Hamiltonian in momentum space in a more familiar form in terms of complex conduction electrons $c_{k,\lambda,\alpha}$ and complex Abrikosov fermions $\chi_{k,\lambda,\alpha}$ from the Kitaev sector (with renormalized coupling constants, $Ku \to K$ and $Ju \to J$).

$$H = \sum_{k,\lambda} \left( \begin{array}{c} c_{k,A,\lambda} \\ c_{k,B,\lambda} \\ f_{k,A,\lambda} \\ f_{k,B,\lambda} \end{array} \right)^\dagger \begin{pmatrix} W & t\theta_k & 0 & 0 \\ \frac{1}{2} & W & 0 & \frac{1}{2} \\ 0 & 0 & K & 0 \\ \frac{1}{2} & 0 & 0 & K \end{pmatrix} \left( \begin{array}{c} c_{k,A,\lambda} \\ c_{k,B,\lambda} \\ f_{k,A,\lambda} \\ f_{k,B,\lambda} \end{array} \right)$$

(3)

where $\theta_k = 1 + e^{-ik_{\mu_1}} + e^{-ik_{\nu_1}}$ and $\lambda = A, B$ takes account of the two equivalent sublattices.

FIG. 1. (a) Optical image of a typical $\alpha$–RuCl$_3$/graphene sample. (b) Schematic illustration of the device, consisting of a hexagonal boron nitride (hBN) capping layer (green) on the top, an $\alpha$–RuCl$_3$ nanosheet (purple) and a graphene monolayer (grey). The temperature-dependent magnetotransport measurements were performed in a similar manner as previously described in Ref. \[25\]. (c) Comparison of the electronic structure from ab-initio calculations \[31\] and the effective low energy model Eq. (1). Inset: Schematic picture of the Kitaev-Kondo lattice and its coupling constants, whereby we considered $K^x = K^y = K^z \equiv K$. 

Hamiltonian reads (dopping all constants),
The Kondo coupling $J$ now appears as an effective hybridisation between the conduction electrons and the formerly fractionalized fermionic excitations of the Kitaev QSL. Note, our aim is not to solve the MF equations numerically but to connect the hFL band structure to the basic features of the ab-initio electronic structure.

Next, we are interested in the low energy limit and expand $\theta_{K+q}$ linearly around momentum $K$ (the contribution from $K'$ is analogous) to obtain

$$H^K = \sum_{q, \sigma} \Phi^K_{q, \sigma} \left( W L + \frac{\sqrt{3}}{2} \frac{t}{\hbar} a^{\sigma} q \frac{j\hbar}{K a^{\tau}} q \right) \Phi^K_{q, \sigma}.$$  \hspace{1cm} (4)

The characteristic energy spectrum of $H^K$, see Fig.1(c), consists of a large Dirac cone from the graphene layer which is shifted by $W$ with respect to the smaller and flat Dirac cone of the Kitaev model. We fix the hopping parameter $t = 2.6$eV by adapting the slope of the graphene Dirac cone to the DFT data. Note, because the Kitaev exchange, $K \approx 17$meV is much smaller than $t$, the Kitaev subsystem has a strongly reduced bandwidth. The large energy shift $W \approx 0.6$eV is in accordance with the charge transfer from graphene to $\alpha$-RuCl$_3$ \cite{39}. Finally, the Fermi energy resides within this correlated layer of the almost flat Dirac cone, giving rise to the hFL behaviour.

We note that in $\alpha$-RuCl$_3$ in proximity to graphene the almost flat Dirac cone originating from the Kitaev model. We fix the hopping parameter $t \approx 17$meV is much smaller than $W_0$ the large energy shift $W_0 \approx 6$eV by adapting the slope of the graphene Dirac cone to the DFT data. Note, because the Kitaev exchange, $K \approx 17$meV is much smaller than $t$, the Kitaev subsystem has a strongly reduced bandwidth. The large energy shift $W \approx 0.6$eV is in accordance with the charge transfer from graphene to $\alpha$-RuCl$_3$ \cite{39}. Finally, the Fermi energy resides within this correlated layer of the almost flat Dirac cone, giving rise to the hFL behaviour.

Remarkably, in the experimentally relevant limit $\mu/J \ll W$ and $\omega_K \to 0$ the LL structure Eq.5 only gives a single pole of the Greens function $G_{\xi,\xi}^{-1}(i\omega_n) = i\omega_n - (E^\xi_{\xi} - \mu)$ given by

$$\Omega_{\text{osc.}} = N_q T \Re \sum_{n=0}^\infty \theta(\Re l_n^*) \sum_{k=1}^\infty \frac{1}{k} e^{i2\pi k l_n^* \text{sgn}(\Im l_n^*)}.$$  \hspace{1cm} (6)

This formula was first reported in Ref. \cite{40}, subsequently employed and benchmarked in Ref. \cite{10}, and derived in its general form in our SM.

Remarkably, in the experimentally relevant limit $\mu/J \ll W$ and $\omega_K \to 0$ the LL structure Eq.5 only gives a single pole of the Greens function

$$l_n^* = \left( \frac{W}{\omega_l} \right)^2 \left( 1 - 2 \frac{i\omega_n}{W} \Gamma \left( \frac{\mu}{J}, \frac{\omega_n}{J} \right) \right),$$  \hspace{1cm} (7)

with $\Gamma\left( \frac{\mu}{J}, \frac{\omega_n}{J} \right) = 1 + \left( \frac{\mu}{J} \right)^2 + \left( \frac{\omega_n}{J} \right)^2 \left( \frac{\mu}{J} \right)^{-1}$. Note, we have checked that setting $\omega_K = 0$ is consistent with a formal, perturbative expansion in $\omega_K/\omega_l \ll 1$. In our approximation we also neglect the term $\frac{\mu}{W} \frac{J^2 l_n^*}{\mu^2 + \omega_n^2}$. 

![FIG. 2. The non-LK behaviour of the QO damping factor \( R(T) \) is plotted as a function of temperature and chemical potential deviation \( \mu/J \). In the hFL regime for small \( \mu/J \) a characteristic maximum appears around \( T_{\text{max}} \approx J/5 \) and at large chemical potential the usual monotonically decreasing hFL behaviour (red, dashed line) is recovered.](image-url)
As our main result, we then obtain the out-of-plane magnetization from the first harmonic \( k = 1 \)

\[
M = -\frac{\partial \Omega_{\text{osc.}}}{\partial B_z} = -\frac{AW}{\varphi_0 \pi} \sin \left( 2\pi \frac{W}{\omega_\text{t}} \right) R(T). \tag{8}
\]

Remarkably, for \( k = 1 \) the magnetization is given by a pure sine-oscillation multiplied by a non-LK damping factor

\[
R(T) = 2\chi \sum_{n=0}^{\infty} e^{-2\chi(n+\frac{1}{2})} \Gamma \left( \frac{\gamma}{2}, \frac{\omega_\text{t}}{2} \right) \quad \text{with} \quad \chi = 4\pi^2 \frac{TW}{\omega_\text{t}^2}. \tag{9}
\]

In contrast to the canonical LK behavior, the damping factor features, for fillings close to the hybridisation region \( \mu \sim J \), a maximum at \( T \approx 1/5 \), see Fig.2.

**Comparison to Experiment.** We have collected QO data from 3 samples. All consist of \( \alpha - \text{RuCl}_3 \) flakes of different thickness on a graphene monolayer, see Fig.1(b). For sample A from Ref. [25] the thickness is 20 nm, while for the new samples B and C it is 20 nm and 4 nm. Note, as graphene is only expected to directly interact with the first \( \alpha - \text{RuCl}_3 \) layers the thickness variation is not expected to strongly influence the charge transport through the proximitized graphene. The temperature-dependent magnetotransport measurements of the samples were performed in a similar manner as previously described in Ref. [25]. In this case we have measured \( R_{xx} \) for \( \pm 4 \) T to \( \pm 12 \) T and various \( T \) from 1 K up to 20 K.

In Fig.3 (a) we show a typical behaviour of the QO at a fixed temperature for sample B (A and C are very similar). We also show our analytically calculated magnetization which shows the same oscillatory behaviour as the experimentally measured longitudinal resistance \( R_{xx} \). We stress that the frequency is in accordance with the charge transfer from ab-initio calculations which was fitted as \( W = 603 \) meV for our effective model in Fig.1(c). To make the comparison of the data to our model quantitative we analysed for each sample the Fourier spectrum as a function of \( B^{-1} \). Each has a sharp maximum in the frequency spectrum which allowed us to extract \( W \) independently from Eq.8. For each sample \( W \approx 600 \) meV within few percent in accordance with the ab-initio fit of our effective model.

Finally, we show the decay of the QO amplitude for increasing temperature in Fig.3 (b). All three samples have a clear non-LK behaviour with samples A, B displaying a clear maximum around \( T_{\text{max}} \approx 7 \) K. We fitted the damping factor Eq.(9) to the experimental data by varying the parameters \( \mu \) and \( J \) (for sample B, C at a fixed \( B \)-field and for sample A from the Fourier transformation of a window around 10 T). We find \( |\mu| < 0.5 \) meV and the key parameter is the Kondo exchange \( J \approx 2 \) meV. Details are given in the SM where we also confirm that we can robustly fit multiple cuts along different \( B \)-fields without changing parameters \( \mu \) and \( J \).

**Discussion.** Our microscopic theory of anomalous QO is based on the hFL electronic structure of the \( \alpha - \text{RuCl}_3 / \) graphene heterostructures and the parameters of the effective model which are determined by ab-initio calculations [24]. The most striking feature of the experimental data is the maximum of the amplitude at a non-zero temperature \( T_{\text{max}} \) which is reproduced in our theory by a Kondo coupling of strength \( J \approx 2 \) meV between the Kitaev honeycomb layer and graphene.

A stringent test of our theory would be a controlled change of the interlayer coupling \( J \), for example via pressure or intercalation, which should lead to a characteristic
shift of the maximum in temperature. Alternatively, because the low energy Kitaev and Kondo scales determine the almost flat band dispersion of the hFL they should be accessible in tunneling measurements, e.g. via scanning tunnelling microscopy (STM).

Previously, the maximum of the amplitude at $\approx 7\, K$ has been tentatively interpreted as a signature of the transition to long-range magnetic order in $\alpha-$RuCl$_3$ [24] because it coincides with its bulk transition temperature $T_N$ [31]. However, we argue that this is unlikely because the diverging magnetic fluctuations upon approaching $T_N$ (from higher or lower temperatures) should lead to an increase in the itinerant electrons scattering rate which then should decrease the QO amplitude at odds with a maximum. In addition, no direct sign of a magnetic transition has been observed in the heterostructure so far.

**Summary and Outlook.** We have developed a theory of *anomalous QO* in $\alpha-$RuCl$_3$ / graphene heterostructures by constructing an effective low energy model which allowed us to derive a new non-LK temperature dependence consistent with our experimental data.

The observation that the frequency and temperature dependence of *anomalous QO* in the Kitaev-Kondo lattice model is consistent with our Shubnikov-de Haas measurements leads to an intriguing interpretation in terms of fractionalized spin excitations within the lattice model [34, 35]. In addition, dynamic fluctuations of the Kitaev QSL acquire charge via the Kondo coupling to the graphene layer and the ensuing hFL band structure gives rise to the *anomalous QO*. Such a scenario directly motivates a search for unconventional superconductivity at lower temperatures which is predicted to occupy a large part of the phase diagram of the Kitaev-Kondo lattice model [41, 33]. In addition, dynamic fluctuations and collective modes of these heterostructures, e.g. the recently observed plasmons [42], are expected to inherit signatures of the proximate QSL.

Our theory of *anomalous QO* will be applicable to other systems which can be described as a Dirac semi-metal in contact with a strongly correlated insulator giving rise to an effective heavy band structure with an almost flat band. The formula for the non-LK temperature dependence can then be turned into a versatile tool to extract the low energy scales of the correlated layer. We expect that other magnetic heterostructures, for example with TaS$_2$ films [13] or the Kagome magnet Nb$_3$X$_8$ (X = Cl, Br) [44], are potential candidates for *anomalous QO*.

On the theory side many questions remain to be explored, for example, how do the fractionalized excitations acquire charge beyond the basic hybridisation picture; or what are alternative microscopic scenarios for QO with charge neutral excitations? Overall, our work paves the way for novel approaches beyond the venerable Lifshitz-Kosevich theory and should also serve as a guide for numerical studies of QO in strongly correlated materials.

**Acknowledgments:** JK thanks K. Burch, N.R. Cooper and E. Henriksen for discussions and collaborations on related work. S.B. and R.V. thank the Deutsche Forschungsgemeinschaft (DFG) for funding through TRR 288-42213477 (A05).

---

[1] L. Onsager, The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science 43, 1006 (1952).
[2] I. Lifshitz and A. Kosevich, Sov. Phys. JETP 2, 636 (1956).
[3] D. Shoenberg, Magnetic oscillations in metals (Cambridge university press, 2009).
[4] L. Taillefer, R. Newbury, G. Lonzarich, Z. Fisk, and J. Smith, in Anomalous Rare Earths and Actinides (Elsevier, 1987) pp. 372-376.
[5] N. Doiron-Leyraud, C. Proust, D. LeBoeuf, J. Levallois, J.-B. Bonnemaison, R. Liang, D. Bonn, W. Hardy, and L. Taillefer, Nature 447, 565 (2007).
[6] B. Tan, Y.-T. Hsu, B. Zeng, M. C. Hatnean, N. Harrison, Z. Zhu, M. Hartstein, M. Kioullappou, A. Srivastava, M. Johannes, et al., Science 349, 287 (2015).
[7] M. Hartstein, W. Toews, Y.-T. Hsu, B. Zeng, X. Chen, M. C. Hatnean, Q. Zhang, S. Nakamura, A. Pudgett, G. Rodway-Gant, et al., Nature Physics 14, 166 (2018).
[8] H. Liu, M. Hartstein, G. J. Wallace, A. J. Davies, M. C. Hatnean, M. D. Johannes, N. Shiitsevalo, G. Balakrishnan, and S. E. Sebastian, Journal of Physics: Condensed Matter 30, 16LT01 (2018).
[9] Z. Xiang, Y. Kasahara, T. Asaba, B. Lawson, C. Tinsman, L. Chen, K. Sugimoto, S. Kagawuchi, Y. Sato, G. Li, et al., Science 362, 65 (2018).
[10] J. Knolle and N. R. Cooper, Phys. Rev. Lett. 115, 146401 (2015).
[11] H. Shen and L. Fu, Physical review letters 121, 026403 (2018).
[12] O. Ertan, P. Ghaemi, and P. Coleman, Physical review letters 116, 046403 (2016).
[13] G. Baskaran, arXiv preprint arXiv:1507.03477 (2015).
[14] D. Chowdhury, I. Sodemann, and T. Senthil, Nature communications 9, 1 (2018).
[15] I. Sodemann, D. Chowdhury, and T. Senthil, Physical Review B 97, 045152 (2018).
[16] L. Zhang, X.-Y. Song, and F. Wang, Physical review letters 116, 046404 (2016).
[17] J. Knolle and N. R. Cooper, Physical Review Letters 118, 176801 (2017).
[18] Z. Han, T. Li, L. Zhang, G. Sullivan, and R.-R. Du, Physical review letters 123, 126803 (2019).
[19] D. Xiao, C.-X. Liu, N. Samarth, and L.-H. Hu, Physical review letters 122, 186802 (2019).
[20] O. I. Motrunich, Physical Review B 73, 155115 (2006).
[21] J. Knolle and R. Moessner, Annual Review of Condensed Matter Physics 10, 451 (2019).
[22] L. Savary and L. Balents, Reports on Progress in Physics 80, 016502 (2016).
[23] K. S. Burch, D. Mandrus, and J.-G. Park, Nature 563, 47 (2018).
[24] B. Zhou, J. Balgley, P. Lampen-Kelley, J.-Q. Yan, D. Mandrus, and E. Henriksen, Physical Review B 100, 165426 (2019).
[25] S. Mashhadi, Y. Kim, J. Kim, D. Weber, T. Taniguchi, K. Watanabe, N. Park, B. Lotsch, J. H. Smet, M. Burghard, and K. Kern, Nano Letters 19, 4659–4665 (2019).
[26] A. Kitaev, Annals of Physics 321, 2–111 (2006).
[27] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, (2016).
[28] M. Hermanss, I. Kimchi, and J. Knolle, Annual Review of Condensed Matter Physics 9, 17 (2018).
[29] S. M. Winter, A. A. Tsirlin, M. Daghofer, J. van den Brink, Y. Singh, P. Gegenwart, and R. Valenti, Journal of Physics: Condensed Matter 29, 493002 (2017).
[30] H. Takagi, T. Takayama, G. Jackeli, G. Khaliullin, and S. E. Nagler, Nature Reviews Physics 1, 264 (2019).
[31] S. Biswas, Y. Li, S. M. Winter, J. Knolle, and R. Valenti, Phys. Rev. Lett. 123, 237201 (2019).
[32] Y. Wang, J. Balgley, E. Gerber, M. Gray, N. Kumar, X. Lu, J.-Q. Yan, A. Fereidouni, R. Basnet, S. J. Yun, et al., arXiv preprint arXiv:2007.06603 (2020).
[33] E. Gerber, Y. Yao, T. A. Arias, and E.-A. Kim, Physical Review Letters 124, 106804 (2020).
[34] U. F. P. Seifert, T. Meng, and M. Vojta, Physical Review B 97 (2018), 10.1103/physrevb.97.085118.
[35] W. Choi, P. W. Klein, A. Rosch, and Y. B. Kim, Physical Review B 98, 155123 (2018).
[36] F. J. Burnell and C. Nayak, Phys. Rev. B 84, 125125 (2011).
[37] R. Schaffer, S. Bhattacharjee, and Y. B. Kim, Phys. Rev. B 86, 224417 (2012).
[38] J. Knolle, S. Bhattacharjee, and R. Moessner, Physical Review B 97, 134432 (2018).
[39] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, Rev. Mod. Phys. 81, 109 (2009).
[40] S. A. Hartnoll and D. M. Hofman, Physical Review B 81 (2010), 10.1103/physrevb.81.155125.
[41] A. Banerjee, P. Lampen-Kelley, J. Knolle, C. Balz, A. A. Aczel, B. Winn, Y. Liu, D. Pajerowski, J. Yan, C. A. Bridges, et al., npj Quantum Materials 3, 1 (2018).
[42] D. J. Rizzo, B. S. Jessen, Z. Sun, F. L. Ruta, J. Zhang, J.-Q. Yan, L. Xian, A. S. McLeod, M. E. Berkowitz, K. Watanabe, et al., arXiv preprint arXiv:2007.07147 (2020).
[43] K. T. Law and P. A. Lee, Proceedings of the National Academy of Sciences 114, 6996 (2017).
[44] C. M. Pasco, I. El Baggari, E. Bianco, L. F. Kourkoutis, and T. M. McQueen, ACS nano 13, 9457 (2019).
Landau level structure

We introduce a magnetic field $B$ over the vector potential $A = (-B_2 y, -B_2 z, -B_k x)^T$ given in the Landau-gauge. Within minimal coupling this can be described by extending the kinetic momentum to the canonical momentum $\hbar q \to \hbar q - eA$. Since we are considering a two dimensional model only the out-of-plane component of the $B$-field couples to momentum. Note that the Landau gauge breaks the translational symmetry in $y$-direction such that $q_y$ is not a good quantum number anymore.

Analogously to determining the Landau levels in graphene, momentum dependent entries correspond to ladder operators of the harmonic oscillator, such that the LL-Hamiltonian reads

$$H^K = \sum_{q_x, \sigma} \Psi^K_{0,q_x,\sigma} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ W & 0 & W & 0 & 0 & 0 & 0 & 0 \\ 0 & W & 0 & W & 0 & 0 & 0 & 0 \\ 0 & 0 & W & 0 & W & 0 & 0 & 0 \\ 0 & 0 & 0 & W & 0 & W & 0 & 0 \\ 0 & 0 & 0 & 0 & W & 0 & W & 0 \\ 0 & 0 & 0 & 0 & 0 & W & 0 & W \\ 0 & 0 & 0 & 0 & 0 & 0 & W & 0 \\ \end{pmatrix} \Psi^K_{0,q_x,\sigma}$$

$$+ \sum_{l=1,q_x,\sigma} \Psi^K_{l,q_x,\sigma} \begin{pmatrix} W & \omega_l \sqrt{I} & 0 & 0 \\ \omega_l \sqrt{I} & W & 0 & 0 \\ 0 & 0 & \omega_l \sqrt{I} & 0 \\ 0 & 0 & \omega_l \sqrt{I} & 0 \end{pmatrix} \Psi^K_{l,q_x,\sigma}$$

(10)

Supplementary Material:

Diagonalizing $[10]$ results in Eq.$[5]$ of the main text where the zeroth LL $l = 0$ has an inherently different form but is not of interested for our current work.

Extending the generalized Lifshitz-Kosevich formula

The goal is to extend the calculation done from (27) to (25) in [40] to poles where $\text{Im} l^* < 0$ or $\text{Re} l^* < 0$. (27) in [40] reads

$$\Omega_{\text{osc.}} = -N_\sigma T \text{Re} \sum_{n=0}^{\infty} \sum_{l=0}^{\infty} \ln(l - l_n^*)$$

(11)

By showing that, up to non-oscillatory terms, the fundamental formula

$$\sum_{l=0}^{\infty} \ln(l - l^*) = -\theta(\text{Re} l^*) \sum_{k=1}^{\infty} \frac{1}{k} e^{2\pi ik^* \text{sgn}(\text{Im} l^*)}$$

(12)

holds, $\Omega_{\text{osc.}}$ can be simplified to $[0]$.

To proof (12) we follow the brief description given in [40]. First we perform a Poisson resummation as given in (14) in [40] and introduce $k \to k + i\epsilon$ with $\epsilon \to 0^+$ to ensure convergence. Then we integrate by parts and split up the sum over $k$.

$$\frac{\sum_{l=0}^{\infty} \ln(l - l^*)}{\ln(x - l^*)e^{2\pi i(k + i\epsilon)x} dx} = \frac{\ln(l^*)}{2\pi \epsilon} + \frac{1}{2\pi \epsilon} \int_{0^-}^{\infty} e^{-2\pi \epsilon x} dx - \frac{\ln(l^*)}{2\pi i} \sum_{k=1}^{\infty} \left( \frac{1}{k + i\epsilon} + \frac{1}{-k + i\epsilon} \right) - \frac{1}{2\pi i} \int_{0^-}^{\infty} \frac{e^{2\pi ikx}}{x - l^*} dx$$

(13)

We have dropped terms which will become large if we take $\epsilon \to 0^+$ but are non-oscillating. The remaining term consists of two exponentials with different signs in the exponent which can be evaluated separately by using complex contour integration. For the first exponential we choose a path $\gamma_{l^*}$ stretching from 0 to $\infty$, then a path $\gamma_{\text{arc}}$ from $\infty$ to $i\infty$ and then along $\gamma_{l^*}$ back to 0. For the second exponential these paths are mirrored at the abscissa.

In both cases we can rewrite, using the residue theorem, the integral along $\gamma_{l^*}$ in terms of the integral along $\gamma_{\text{arc}}$ and a contribution of the poles in the first (+) or fourth (−) quadrant. Since the integrand vanishes for $x \to \infty$ the integral along $\gamma_{\text{arc}}$ vanishes. Furthermore the integral along $\gamma_{l^*}$ is non-oscillatory. Therefore the only oscillatory contribution of the integral comes from the residue of the poles, located in the right half plane

$$\gamma_{\text{arc}}$$ and $\gamma_{l^*}$ and then along $\gamma_{l^*}$ back to 0. For the second exponential these paths are mirrored at the abscissa.

In both cases we can rewrite, using the residue theorem, the integral along $\gamma_{l^*}$ in terms of the integral along $\gamma_{\text{arc}}$ and a contribution of the poles in the first (+) or fourth (−) quadrant. Since the integrand vanishes for $x \to \infty$ the integral along $\gamma_{\text{arc}}$ vanishes. Furthermore the integral along $\gamma_{l^*}$ is non-oscillatory. Therefore the only oscillatory contribution of the integral comes from the residue of the poles, located in the right half plane

$$\frac{1}{2\pi i} \int_{0^-}^{\infty} \frac{e^{2\pi ikx}}{x - l^*} dx = \frac{1}{k} \theta(\text{Re} l^*) e^{2\pi i k^* \text{sgn}(\text{Im} l^*)}$$

(14)
Fitting of experimental data at different $B$-fields

FIG. 4. Fit of various amplitudes at different $B$-fields for sample B, predicting $|\mu| = 0.41$ meV and $J = 1.71$ meV.

| sample | A  | B  | C  | B for various $B$s |
|--------|----|----|----|------------------|
| $f_{B-1}$ | 365 T | 390 T | 384 T | 390 T |
| $B_z$ | 10 T | 11.7 T | 9.82 T | 8/9.5/11/11.7 T |
| $W$ | 583 meV | 603 meV | 598 meV | 603 meV |
| $\omega_t$ | 96.5 meV | 104 meV | 95.7 meV | 86/94/101/104 meV |
| $J$ | 2.30 meV | 1.78 meV | 2.82 meV | 1.70 meV |
| $|\mu|$ | 271 $\mu$eV | 324 $\mu$eV | 520 $\mu$eV | 407 $\mu$eV |

TABLE I. Fitting parameters of the curves in Fig.3 (b) (first three columns) and Fig.4 (last column). $W$ and $\omega_t$ are calculated from the magnetic field $B$ and the frequency $f$ which are determined from the experiment, $J$ and $\mu$ are fitting parameters.