Excitons in topological Kondo insulators – theory of thermodynamic and transport anomalies in SmB_6

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Kondo insulating materials lie outside the usual dichotomy of weakly versus correlated – band versus Mott – insulators. They are metallic at high temperatures but resemble band insulators at low temperatures because of the opening of an interaction induced band gap. The first discovered Kondo insulator (KI) SmB_6 has been predicted to form a topological KI (TKI). However, since its discovery thermodynamic and transport anomalies have been observed that have defied a theoretical explanation. Enigmatic signatures of collective modes inside the charge gap are seen in specific heat, thermal transport and quantum oscillation experiments in strong magnetic fields. Here, we show that TKIs are susceptible to the formation of excitons and magneto-excitons. These charge neutral composite particles can account for long-standing anomalies in SmB_6.

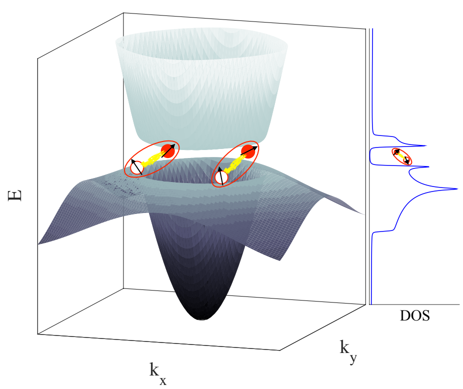

FIG. 1. At low temperatures strong correlations lead to the formation of fermionic quasiparticles which have the schematically shown band structure (for $k_z = 0$) of a broad-brimmed Mexican hat. Due to the large DOS at the band edges it is very susceptible to the formation of excitons.

One of the biggest successes of quantum mechanics is the explanation of the distinction between metals and insulators. Traditionally, there are two different regimes: First, in weakly interacting systems insulating behaviour arises from complete filling of Bloch bands with a gap to unoccupied states, whereas metals have partially filled bands giving a manifold of gapless excitations – the Fermi surface (FS). Second, in strongly interacting systems repulsion forbids hopping of electrons leading to Mott insulators. However, a third possibility exists – so called Kondo insulators (KI) – where strong interactions between itinerant electrons and localized spins lead to a heavy band insulator at low temperatures [1].

The material SmB_6 was the first KI discovered almost half a century ago [2]. More recently, it has been predicted that SmB_6 should be a topological KI (TKI) [3] which is an interaction-induced heavy 3D topological insulator [4]. Normally the charge gap in an insulator also determines its thermodynamic and bulk transport properties which are expected to then follow an Arrhenius type activated temperature dependence. However, the tentative TKI SmB_6 strongly deviates from that picture and exhibits unusual thermodynamic anomalies, for example a low temperature specific heat contribution reminiscent of a metal [5, 6]. These have not found a generally accepted explanation for decades and more recent experiments motivated from the TKI proposal have added even bigger puzzles. The observation of bulk quantum oscillations (QO) [7], normally a synonym for a FS, and residual thermal transport inside the insulating low temperature regime [8] challenge our canonical understanding of metals and insulators.

Here, we show TKIs are very susceptible to the formation of *excitons* or *magneto-excitons* (MExc) in an applied magnetic field B. The strong Coulomb repulsion of the localised Sm f-levels has two effects: First, it gives rise to a heavy insulating state with a peculiar broad-brimmed Mexican hat-like band structure, see Fig.1, which provides the necessary large density of states (DOS) from the band extrema, see right panel. Second, it provides the interaction which binds the particle-hole pairs. While spin-excitons[20–22] have been discussed in connection with collective in-gap modes observed in inelastic neutron scattering (INS) [17–19] before, here we show that a new exciton branch at lower energies can provide a natural explanation of long standing thermodynamic anomalies in SmB_6. In addition, by generalising the theory of MExc to TKIs we discover a new mechanism by which an insulator without a FS can exhibit bulk quantum oscillations.

The model. We focus on a minimal model of a TKI which captures the essential physics of SmB_6 and takes the form of a periodic Anderson lattice model [3, 9].

$$H = \sum_{\mathbf{k},\alpha\beta} \begin{pmatrix} d^\dagger_{\mathbf{k},\alpha} & f^\dagger_{\mathbf{k},\alpha} \end{pmatrix} \begin{pmatrix} \epsilon^d_{\mathbf{k}} & \frac{\gamma}{2}\vec{s}_{\mathbf{k}}\vec{\sigma}_{\alpha,\beta} \\ \frac{\gamma}{2}\vec{s}_{\mathbf{k}}\vec{\sigma}_{\alpha,\beta} & \epsilon^f_{\mathbf{k}} \end{pmatrix} \begin{pmatrix} d_{\mathbf{k},\beta} \\ f_{\mathbf{k},\beta} \end{pmatrix} + U \sum_i f^\dagger_{\mathbf{r}_i,\uparrow} f_{\mathbf{r}_i,\uparrow} f^\dagger_{\mathbf{r}_i,\downarrow} f_{\mathbf{r}_i,\downarrow} \quad (1)$$

The broad Sm d-band has a dispersion $\epsilon^d_{\mathbf{k}}$ =

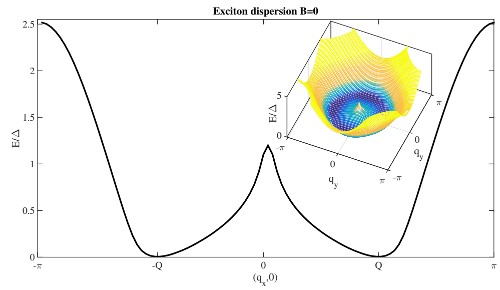

FIG. 2. Shown for $B = 0$ as a function of q_x with $q_y = 0$. The full exciton dispersion of the 2D TKI is plotted in the inset. Because of the special band structure the dispersion minima are always at a nonzero momenta but their precise energy depends on the interaction strength. We have used parameters $W = \mu = -3.2$, $\alpha = 0.15$, $\gamma = 0.1$, $U = 2.75$ (in units of hopping t), which give a dispersion with a small gap which can best explain experiments on SmB_6.

$-2t\sum_{\mathbf{k},\eta=x,y,z}\cos k_\eta$. The almost flat inverted f-band is $\epsilon^f_{\mathbf{k}} = -\alpha\epsilon^d_{\mathbf{k}} + \lambda$ (with $\alpha \ll 1$) with $\lambda = (1+\alpha)W$ and incorporates the strong on-site repulsion U. The d- and f-level have different angular momenta and the latter is a superposition of different spin components due to strong spin orbit coupling. Hence, the hybridisation in a TKI is spin and momentum-dependent (odd-parity), $\vec{s}_{\mathbf{k}} = (\sin k_x, \sin k_y, \sin k_z)^{\mathrm{T}}$.

To account for the strongly correlated nature we treat the self energy of the f-electrons as momentum independent such that the main effect of the local repulsion is a renormalisation of the f-bands and the hybridisation [10]. The low energy excitations are described by a new non-interacting Hamiltonian identical to the first term in Eq.1 but with new parameters $\tilde{t}$ and $\tilde{\gamma}$ [11, 12] (we drop the tilde in the remainder). Here, we use effective parameters in agreement with the experimentally known dispersion around the three X-points of SmB_6 [13–15].

The Hamiltonian at low temperatures yields two two-fold degenerate energies $E^\nu_\pm(\mathbf{k}) = \frac{1}{2}\left[\epsilon^d_{\mathbf{k}} + \epsilon^f_{\mathbf{k}}\right] \pm d_{\mathbf{k}}$ with $d_{\mathbf{k}} = \frac{1}{2}\sqrt{\left(\epsilon^d_{\mathbf{k}} - \epsilon^f_{\mathbf{k}}\right)^2 + |\gamma s_{\mathbf{k}}|^2}$. A schematic form of the band structure is shown in Fig.1. In the TKI state the lower bands are completely filled and the broad-brimmed Mexican hat-like dispersion gives a large DOS near the gap. For simplicity, we concentrate in the following on an effective two-dimensional model but we have checked that the inclusion of the third dimension does not lead to any qualitative changes of our findings. Setting $k_z = 0$ the Hamiltonian decouples into two independent blocks, labeled by $\nu = +1(-1)$, for the $d^\dagger_\uparrow f_\downarrow$ and $d^\dagger_\downarrow f_\uparrow$ states diagonalised by

$$c_{\mathbf{k},\nu} = \cos\frac{\beta_{\mathbf{k}}}{2} d_{\mathbf{k},\nu} + \sin\frac{\beta_{\mathbf{k}}}{2} e^{\mathrm{i}\nu\theta_{\mathbf{k}}} f_{\mathbf{k},\bar{\nu}} \qquad (2)$$

$$v_{\mathbf{k},\nu} = -\sin\frac{\beta_{\mathbf{k}}}{2} e^{-\mathrm{i}\nu\theta_{\mathbf{k}}} d_{\mathbf{k},\nu} + \cos\frac{\beta_{\mathbf{k}}}{2} f_{\mathbf{k},\bar{\nu}}$$

with the angles given by $\cos\beta_{\mathbf{k}} = \frac{1}{2d_{\mathbf{k}}}\left[\epsilon^d_{\mathbf{k}} - \epsilon^f_{\mathbf{k}}\right]$ and $\sin\beta_{\mathbf{k}} e^{-\mathrm{i}\theta_{\mathbf{k}}} = \frac{\gamma}{2d_{\mathbf{k}}}(\sin k_x - \mathrm{i}\sin k_y)$.

Excitons. To investigate the formation of bound excitons, we include the strong f-level interaction on top of these bands. We project the Hubbard term of Eq.1 onto the renormalised TKI bands and concentrate only on those terms which lead to exciton binding

$$H_{\text{int}} = -U\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}}\Big[\phi(\mathbf{k}+\mathbf{q},-\mathbf{q})\phi^*(\mathbf{k}'+\mathbf{q},-\mathbf{q})c^\dagger_{\mathbf{k}+\mathbf{q},-}v_{\mathbf{k},+}v^\dagger_{\mathbf{k}',+}c_{\mathbf{k}'+\mathbf{q},-} + \phi(\mathbf{k},\mathbf{q})\phi^*(\mathbf{k}',\mathbf{q})v^\dagger_{\mathbf{k}+\mathbf{q},-}c_{\mathbf{k},+}c^\dagger_{\mathbf{k}',+}v_{\mathbf{k}'+\mathbf{q},-}\Big] \qquad (3)$$

with $\phi(\mathbf{k},\mathbf{q}) = \sin\frac{\beta_{\mathbf{k}}}{2}\cos\frac{\beta_{\mathbf{k}+\mathbf{q}}}{2}e^{-\mathrm{i}\theta_{\mathbf{k}}}$. Hence, our system is described by the Hamiltonian $H = H_0 + H_{\text{int}}$ with $H_0 = \sum_{\mathbf{k},\nu}\left[E^\nu_+(\mathbf{k})c^\dagger_{\mathbf{k},\nu}c_{\mathbf{k},\nu} + E^\nu_-(\mathbf{k})v^\dagger_{\mathbf{k},\nu}v_{\mathbf{k},\nu}\right]$ and the 'valence' and 'conduction' bands E^ν_- and E^ν_+ separated by a gap $\Delta = \min(E^\nu_+) - \max(E^\nu_-)$. The interaction only binds electron-hole pairs of opposite ν. For example, $(-\,+)$-excitons [similarly for the time reversed partner $(+\,-)$] are created by the operator

$$S^\dagger_{\mathbf{q}} = \sum_{\mathbf{k}}\varphi_{\mathbf{q}}(\mathbf{k})c^\dagger_{\mathbf{q},-}v_{\mathbf{k}+\mathbf{q},+} \qquad (4)$$

which are directly related to spin flip excitations. We calculate their dispersion $E(\mathbf{q})$ from the Bethe-Salpeter equation

$$\left[H_0 + H_{\text{int}}, S^\dagger_{\mathbf{q}}\right]|0\rangle = E(\mathbf{q})S^\dagger_{\mathbf{q}}|0\rangle. \qquad (5)$$

We evaluate the quartic operators from the interaction within the ground state, $v^\dagger_{\mathbf{k}}|0\rangle = 0$ and $c_{\mathbf{k}}|0\rangle = 0$, which is equivalent to an RPA-type diagrammatic treatment [16]. We obtain a non-linear equation for the exciton wave function $\varphi_{\mathbf{q}}(\mathbf{k})$ and its dispersion $E(\mathbf{q})$. Due to the fact that the interaction factorises, as $U\phi(\mathbf{k},\mathbf{q})\phi^*(\mathbf{k}',\mathbf{q})$, this can be cast as a simple implicit equation for $E(\mathbf{q})$

$$1 = -U\sum_{\mathbf{k}}\frac{|\phi(\mathbf{k},\mathbf{q})|^2}{E(\mathbf{q}) - \left[E^-_+(\mathbf{k}+\mathbf{q}) - E^+_-(\mathbf{k})\right]}. \qquad (6)$$

We show a representative exciton dispersion in the upper panel of Fig.2. It has an almost degenerate ring-like manifold of energy minima at momenta $\mathbf{Q}$, see inset. This peculiar form of the dispersion originates from the special form of the TKI band structure where $\mathbf{Q}/2$ are the vectors pointing to the maxima (minima) of the bands.

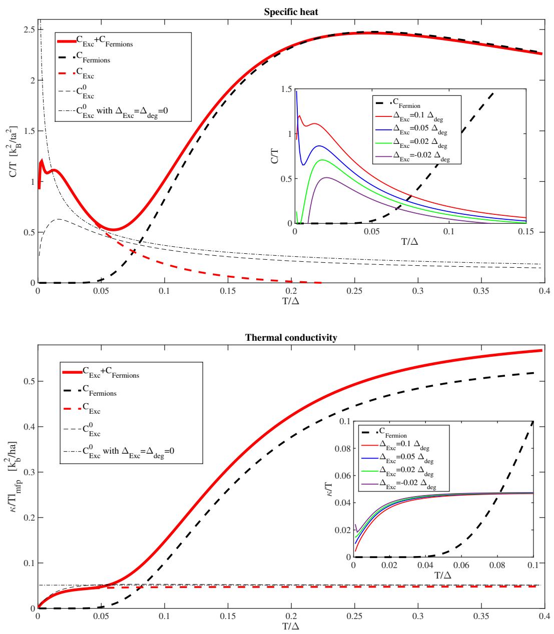

FIG. 3. The upper panel shows the specific heat for $B = 0$, plotted as C/T, as a function of scaled temperature T/Δ with the fermionic band gap Δ. For comparison, we plot the behaviour of non-interacting excitons, $C^0_{\rm Exc}$, with a dispersion with a perfect degenerate ring-like minimum (black dot dashed) or in the self-consistent lattice dispersion (black dashed). We use the same parameters as in the upper panel of Fig.3 (with $\Delta_{\rm deg}/\Delta = 1/20$, $\Delta_{\rm Exc} = \Delta_{\rm deg}/10$, $g = 0.004t$) and scaled the exciton contribution by a factor 1/4. The lower panel shows the thermal conductivity plotted as κ/T.

In order to account for the anomalies in SmB_6 the exciton binding energy needs to be large such that the exciton gap $\Delta_{\rm Exc}$ is small compared to the bandgap and sets a low temperature scale. In the simplest approximation excitons can be treated as a non-interacting gas of bosons, with specific heat $C = \sum_{\mathbf{k}} E(\mathbf{k}) \frac{\partial n_{\mathbf{k}}}{\partial T}$ where $n_{\mathbf{k}} = \left[e^{\frac{E(\mathbf{k})}{k_B T}} - 1 \right]^{-1}$. The almost degenerate ring-like minimum of the excitons has strong consequences for the behaviour of experimental observables because it reduces the effective dimensionality to one dimension. For perfect degeneracy and $\Delta_{\rm Exc} = 0$ such that $E(\mathbf{q}) \propto (|\mathbf{q}| - |\mathbf{Q}|)^2$, it is easy to show that a residual specific heat contribution $C/T \propto 1/\sqrt{T}$ appears independent of dimensionality!

Although it might appear that one must tune $\Delta_{\rm Exc} \approx 0$ to obtain this interesting low-temperature behaviour, in fact no such fine-tuning is needed in more realistic models. First, lattice effects lift the degeneracy such that the divergence in specific heat would be removed below a scale $\Delta_{\rm deg}$ which is the difference between the maximum and minimum energy on the ring. Second, Pauli blocking suppresses the formation of excitons with similar momenta. Third, exciton-exciton interactions should be included, and suppress the exciton number. We have modelled the influence of these effects on $n_{\mathbf{k}}$, including exciton-exicton interactions in a self-consistent theory (details are in the supplementary material [35]). The qualitative low-temperature behaviour of the free boson approximation survives without any fine-tuning of parameters, e.g. even for a small negative gap the exciton density remains small.

The resulting specific heat is shown in the upper panel of Fig.3. Clearly, excitons inside the band gap lead to an extra contribution in contrast to a pure fermionic scenario (black dashed) which has exponential suppression at low T [6]. The main panel shows the specific heat as calculated from the self-consistently determined exciton dispersion compared to the asymptotic behaviour of a non-interacting model with $\Delta_{\rm deg} = 0$ (thin dot dashed) and $\neq 0$ (thin dashed). In the inset of Fig.3 we show C/T calculated for different values of the exciton gap. We find a strong low-temperature exciton contribution with an upturn very similar to experiments on SmB_6 [5, 6].

Charge neutral excitons cannot lead to charge transport, however, they can conduct heat. Within a semiclassical Boltzmann-like treatment we calculate their thermal conductivity as $\kappa = \sum_{\mathbf{k}} l_{\rm mfp} |v_{\mathbf{k}}| E(\mathbf{k}) \frac{\partial n_{\mathbf{k}}}{\partial T}$ with velocity $v_{\mathbf{k}} = \frac{\partial E(\mathbf{k})}{\partial \mathbf{k}}$ and the mean free path $l_{\rm mfp}$. Excitons dominate thermal conductivity at temperatures below the charge gap. Their contribution originates again from the special form of the exciton dispersion, e.g. non-interacting bosons with a gapless dispersion with a perfectly degenerate ring-like minimum directly give $\kappa/T =$ const., thus, mimicking the behaviour of a metal. As shown in the lower panel of Fig.3, in our interacting lattice calculation this linear κ-term is non-zero but strongly reduced for increasing $\Delta_{\rm Exc}$, see inset. Since the precise value of the exciton gap is sensitive to microscopic details, there may be variations in the asymptotic low temperature behaviour between samples [8, 28] arising from small changes in this gap.

Magneto-excitons for $B > 0$. Already a weak field has direct consequences in our scenario: e.g. the lifting of degenerate exciton branches ($c^\dagger_- v_+$ and $c^\dagger_+ v_-$) from a Zeeman coupling, and a reduction of the fermionic gap which leads to an increase of the thermal conductivity κ and the specific heat anomaly [29].

But could excitons also lead to a de Haas van Alphen effect (dHvAe)? Of course, the MExc itself is charge neutral and its center of mass does not undergo cyclotron motion due to the absence of a Lorentz force [30]. However, they are built of particle and hole bands that form discrete Landau levels (LLs) in a magnetic field which in turn can lead to QO even for band insulators [31, 32]. Hence, the energy of the MExc varies periodically as a function of $1/B$ through the variation of the energy of its constituents. This ultimately simple mechanics (abbreviated as EdHvAe) leads to an oscillatory behaviour of the total energy as a function of $1/B$ and hence the

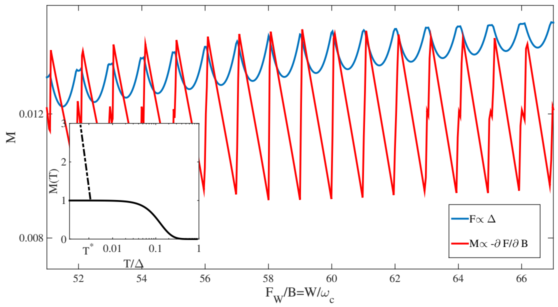

FIG. 4. The energy of MExc, which is mainly given by the LL gap for weak MExc binding, changes periodically as a function of inverse magnetic field, see Eq.7. Therefore, the free energy of the exciton system F (blue) varies periodically together with the corresponding magnetisation M (red) (same parameters as before and $\gamma/W = 0.025$). The schematic temperature dependence is shown in the inset.

magnetisation.

The temperature dependence of the EdHvAe can be estimated qualitatively. We have shown recently [31] that narrow gap insulators can exhibit an *anomalous* de Haas van Alphen effect (AdHvAe) if the cyclotron frequency $\hbar\omega_c$ is comparable to the activation gap of the insulating state, see also Ref.[32]. SmB_6 has an activation gap of about $\approx$ 10 meV [15, 19] and a small effective mass of the itinerant d-band [13, 15], $m_d = 0.44 m_e$, leading to $\hbar\omega_c \approx$ 4meV for 15 Tesla. Hence, the $B = 0$ gap is slightly bigger than the cyclotron energy, however, in a TKI the gap is reduced in a magnetic field [32]. Overall, we expect that the amplitude of oscillations from the AdHvAe is small but otherwise follows a Lifshitz-Kosevich temperature dependence [31, 32] at higher temperatures. Since the EdHvAe also originates from the periodic variation of gapped LL branches we expect a temperature dependence similar to that of the AdHvAe but with an enhanced intensity.

It is tempting to speculate about the low temperature contribution of MExc which is complicated by the appearance of several low energy scales, e.g $\Delta_{\text{deg}}, \Delta_{\text{Exc}}$. For $\Delta_{\text{Exc}} < 0$ a condensate should form at a temperature T^* below which the density of MExc quickly grows leading to a steeply increasing QO amplitude. A schematic plot of such a temperature dependence is shown in the inset of Fig.4. For $\Delta_{\text{Exc}} > 0$ we would expect a thermally activated number of excitons (and contribution to QO) for $T < \Delta_{\text{Exc}}$. However, when projecting the f-level repulsion onto the TKI bands we have only concentrated on those terms which lead to direct exciton binding, but there exist residual terms which create or destroy pairs of $(+-)$ and $(-+)$ excitons. On the one hand, these exciton-non-conserving terms smoothen out the condensation transition to a mere crossover. On the other hand, they lead to a correction to the ground state energy from virtual exciton-pair fluctuations which entails a contribution to the QO amplitude even in the absence of thermally excited excitons for $\Delta_{\text{Exc}} > 0$ and $T \to 0$.

We can corroborate our qualitative discussion of the EdHvAe by a microscopic calculation in the strong field limit. Details of our MExc calculation from, Eq.1, are in the supplementary material [35]. Similar to the $B = 0$ case, the interaction only binds particle-hole pairs from opposite sectors ν. For a given magnetic field there is a LL index N_- (N_+) such that the energy of the corresponding valence (conduction) LL branch $E^{\nu}_-(N_-)$ ($E^{\bar{\nu}}_+(N_+)$) is maximal (minimal). We study the formation of excitons between those extremal levels and obtain a closed expression for the ME dispersion

$$E(\mathbf{k}) = \Delta(B) - U\left[\cos\frac{\beta^-_{N_+}}{2}\right]^2\left[\sin\frac{\beta^+_{N_-}}{2}\right]^2 \frac{(-1)^{N_-+N_+-1}}{2\pi} e^{-\frac{k^2}{2}} L^{N_--N_++1}_{N_+-1}\left(\frac{k^2}{2}\right) L^{N_+-1-N_-}_{N_-}\left(\frac{k^2}{2}\right) \qquad (7)$$

with the LL gap $\Delta(B) = E^-_+(N_+) - E^+_-(N_-)$ and the angles given by $\tan\beta^{\nu}_n = \frac{\hbar\gamma}{2\sqrt{2}l_B}\frac{\sqrt{n}}{\hbar\omega_c\left(n-\nu\frac{1}{2}\right)+\alpha\hbar\omega_c\left(n+\nu\frac{1}{2}\right)-\lambda}$ ($l_B = \sqrt{\frac{c\hbar}{eB}}, \omega_c = \frac{eB}{m_d c}$).

In the regime $E_B < \hbar\omega_c < \Delta(B)$ the change of the exciton dispersion $E(\mathbf{k})$ as a function of field arises mainly from the changing LL gap $\Delta(B)$, see Eq.7. Then, for a given temperature the free energy and the magnetisation are directly related to the periodic variation of the gap $M = -\frac{\partial F}{\partial B} \propto -\frac{\partial \Delta}{\partial B}$. In Fig.4 we plot this oscillatory behaviour as a function of $W/\hbar\omega_c = F_W/B$. There is indeed an exciton contribution to the magnetisation whose period $1/F_W$ corresponds to a dHvAe frequency set by the area of the intersection of the unhybridised bands similar to experimens in SmB_6 [7].

Discussion. Several experiments on SmB_6 have observed in-gap states reminiscent of our excitons. INS measured weakly dispersing 'spin-excitons' [17–22] but with a much bigger gap. Scanning tunneling spectroscopy [23] and inelastic light scattering [24] find evidence for collective modes. The notorious resistivity plateau at low temperatures, which was originally attributed to impurity bands [25], has also been related to exciton-complexes which acquire charge by trapping electrons [26]. However, transport measurements [27] can relate the plateau to surface states which invalidates such a polaronic explanation.

In our excitonic scenario we expect that applying a magnetic field should increase C, κ and T^*. To settle the ongoing debate whether the experimentally observed QOs are coming from gapless surface states [30, 33, 34], or from the bulk [7, 31, 32] one should directly compare the field dependence of charge transport [Shubnikov de Haas effect (SdHe)], and thermodynamic quantities (dHvAe). If both were only due to gapless surface states the behaviours of SdHe and dHvAe should be similar to those in a metal. However, if one is a bulk signal and the other from the surface we expect that both differ strongly.

There are strong predictions to test our scenario: First, despite the absence of a bulk SdHe there should be QO in the *thermal* conductivity which would also allow a clear separation of the exciton contribution from phonons. Second, in addition to the observed spin-excitons around 10 meV [17–19] we expect another very low energy mode in INS experiments $\lesssim 1$ meV, which so far could have been masked by the quasi-elastic signal in previous experiments which focused on energies above 5meV.

In conclusion, we have shown that TKIs are susceptible to the formation of excitons with a ring-like dispersion. They gives rise to a low temperature specific heat contribution, residual thermal conductivity and an unexpected dHvAe in agreement with observations in SmB_6.

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