Anomalous de Haas–van Alphen Effect in InAs/GaSb Quantum Wells

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The de Haas–van Alphen effect describes the periodic oscillation of the magnetization in a material as a function of an inverse applied magnetic field. It forms the basis of a well established procedure for measuring Fermi surface properties, and its observation is typically taken as a direct signature of a system being metallic. However, certain insulators can show similar oscillations of the magnetization from quantization of the energies of electron states in filled bands. Recently, the theory of such an *anomalous* dHvAE (AdHvAE) was worked out, but there has not yet been a clear experimental observation. Here, we show that the inverted narrow gap regime of InAs/GaSb quantum wells is an ideal platform for the observation of the AdHvAE. From our microscopic calculations, we make quantitative predictions for the relevant magnetic field and temperature regimes, and we describe unambiguous experimental signatures.

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Introduction.—In his seminal paper on the diamagnetism of metals in 1930 [1], Landau discovered what are now known as *quantum oscillations* (QOs), which describe the periodic variation of experimental observables as a function of the applied magnetic field B. However, unaware of the experimental discovery in the same year of QO in the magnetization [2]—the de Haas–van Alphen effect (dHvAE)—as well as the conductivity [3]—the Shubnikov–de Haas effect (SdHE)—Landau erroneously dismissed the effect as unobservably small. The subsequent work of Onsager [4] and Lifshitz-Kosevich (LK) [5] showed the direct connection of the period of QOs to extremal areas of Fermi surfaces (FSs) and of the temperature dependence of the amplitude to the effective mass of the electrons. This quantitative theory has turned the effect into the most precise experimental tool for measuring FS properties [6] now in standard use around the world.

The dHvAE and SdHE originate from the periodic crossing of quantized Landau levels (LLs) through the chemical potential, such that measuring these effects in a material is almost taken as a synonym for the system being metallic [6]. Therefore, the observation of the dHvAE in the insulating state of the heavy-fermion material SmB_6 [7] came as a big surprise. Motivated by this experiment, we recently showed that, contrary to common intuition, QOs can appear in certain band insulators [8]—dubbed the *anomalous* dHvAE (AdHvAE). The prerequisites are that the filled and empty bands should be separated by a hybridization gap that is on the order of the relevant cyclotron frequency, $\hbar\omega_c$, and that this hybridized region should trace out a well-defined closed surface in momentum space (a "shadow FS") at which the dispersion of the filled band changes abruptly. (In the cases described below, this will be at the maximum of the filled band $E_{\max}$.) For a given magnetic field B, this results in a LL structure with a sharp change in its dispersion as a function of LL index n at the energy $E_{\max}$. The distance between LLs set by $\hbar\omega_c \propto B$ changes as a function of the field such that subsequent levels are pushed over $E_{\max}$ [9], causing the thermodynamic potential to oscillate even in an insulator. The frequency of oscillations $F = \hbar A_S/2\pi e$ is set by the area A_S traced out by the momenta corresponding to energy $E_{\max}$ (the area of the shadow FS).

Our previous work [8] was based on a flat band of f electrons hybridized with a dispersive band of d electrons. The connection of such a simple noninteracting toy model to the complicated Kondo insulator SmB_6 is currently under debate [10–13]. However, subsequently, the AdHvAE was shown to occur in other insulating band structures

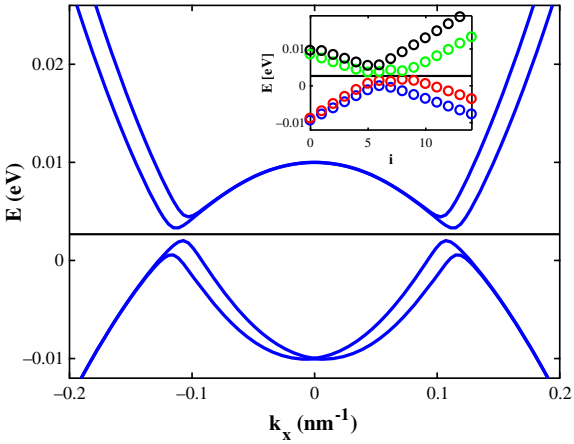

FIG. 1. The low energy band structure of InAs/GaSb QWs are shown for a system close to the gap-closing transition with a small gap of $\Delta = 1.3$ meV. We assume that the chemical potential is in the middle of the gap (the solid line). (Inset) The calculated Landau level structure for $B = 0.6$ T with upward and downward dispersing LL branches, as a function of an index $i = 0, 1, 2, \ldots$ labeling the states in each band.

fulfilling the abovementioned prerequisites. Model calculations for topological insulators [9] and gapped semimetals [14] have explored the effect in more general settings, but an unambiguous experimental observation of the ultimately elementary AdHvAE is missing.

Here, we address the following outstanding question: What is a realistic and readily available experimental system for observing the AdHvAE? Is it possible to make quantitative predictions? We propose that the insulating regime of an electron hole bilayer with a small hybridization gap is the system of choice. Specifically, InAs/GaSb quantum wells (QWs) [15] constitute an ideal platform because they combine a number of desirable features: (i) They have an inverted band structure resulting in upward and downward dispersing LLs; see Fig. 1, whose intersection provides a well-defined closed contour in momentum space. (ii) Their dispersion and the band gap are highly tunable, as confirmed by a recent observation of the metal-insulator transition [16,17]. (iii) They are well described by effective noninteracting models [15]. (iv) They have small band masses resulting in sizable cyclotron frequencies for small magnetic fields, such that the AdHvAE should be observable in a broad regime of band gaps. (v) In contrast to HgTe QWs [18], which are very difficult to fabricate [19], InAs/GaSb QWs are much simpler, with many different groups studying the quantum spin Hall properties [16,20–25].

By performing a first quantitative calculation, we show that the AdHvAE is straightforwardly accessible in InAs/GaSb QWs for magnetic fields below 2 T. A direct observation of QO in the magnetization and the simultaneous absence of QO in the conductivity (no SdHE) will be a smoking gun signature of its discovery.

Effective four-band model.—The low energy electronic degrees of freedom can be described by an effective four-band model in the basis of electron- or holelike (e-h) states $[e\uparrow, h\uparrow, e\downarrow, h\downarrow]$ [15]. The Hamiltonian, which is similar to the Bernevig-Hughes-Zhang model describing the quantum spin Hall effect in HgTe/CdTe QWs, is described by [15,26]

$$\hat{H} = \begin{pmatrix} M_0 + \mu_+ k^2 - \frac{g_e \mu_B}{2} B & \beta k_+ & \Delta_+ k_+ - i\alpha k_- & -\Delta_0 \\ \beta k_- & -M_0 - \mu_- k^2 - \frac{g_h \mu_B}{2} B & \Delta_0 & \Delta_- k_- \\ \Delta_+ k_- + i\alpha k_+ & \Delta_0 & M_0 + \mu_+ k^2 + \frac{g_e \mu_B}{2} B & -\beta k_- \\ -\Delta_0 & \Delta_- k_+ & -\beta k_+ & -M_0 - \mu_- k^2 + \frac{g_h \mu_B}{2} B \end{pmatrix}, \quad (1)$$

with $k_+ = (k_x + ik_y)$, $k_- = k_+^*$, $k^2 = k_x^2 + k_y^2$. The parameter β controls the degree of hybridization between the subbands systems, thus largely determining the size of the hybridization gap. In actual materials, it can be widely controlled by, for example, varying layer thickness or backgating [16,20,27].

The terms proportional to Δ_i ($i = +, -, 0$) describe the bulk inversion asymmetry (BIA), and α terms arise from the structural inversion symmetry (SIA). In contrast to HgTe/CdTe QWs, the electron and hole subbands are localized in spatially separated layers in InAs/GaSb QWs. Therefore, the inversion symmetry is broken and the SIA terms dominate over the BIA terms. Since the latter are at least an order of magnitude smaller, we can safely neglect them ($\Delta_i = 0$ in the following), which facilitates the calculation of the quantized LL dispersion in an orbital magnetic field. Finally, we have included a Zeeman term which will lift some of the remaining spin degeneracies. Typical values have been determined very recently, and they are around $g_e \approx 10$ and $g_h \approx 3$ [23]. In Fig. 1 we show the band structure for a QW structure close to the critical thickness with a small band gap of 1.3 meV. The microscopic parameters have been calculated previously and we use the same values as Ref. [28], taken from Ref. [26] except for a smaller β [0.12 eV Å], which reduces the gap bringing the system closer to the metal-insulator transition; see also the Supplemental Material [29].

In the following we will set up the calculation for the energy levels in an orbital magnetic field [27,30,31] which can then be used to directly calculate experimental observables. We introduce B in the out of plane direction via the vector potential $\mathbf{A}$, which is minimally coupled to the crystal momentum such that $\mathbf{\Pi} = \hbar\mathbf{k} + (e/c)\mathbf{A}$. Then we can replace the momentum operators in Eq. (1) with the standard ladder operators $k_+ \to (\sqrt{2}/l_B)\hat{a}^\dagger$, $k_- \to (\sqrt{2}/l_B)\hat{a}$, and $k^2 \to (2/l_B^2)(\hat{a}^\dagger\hat{a} + \frac{1}{2})$, with the magnetic length $l_B = \sqrt{\hbar c/e|B|} \approx 26\ \mathrm{nm}/\sqrt{B(\mathrm{T})}$. With the ansatz wave function $|\Psi_n\rangle = [u_n|n\rangle, v_n|n-1\rangle, w_n|n+1\rangle, x_n|n+2\rangle]^T$ in terms of the standard harmonic oscillator states $|n>$, the problem of calculating the energy levels, $\hat{H}|\Psi_n\rangle = E_n|\Psi_n\rangle$, for a given field B as a function of the LL index n is reduced to that of finding the eigenvalues of

$$H_n = \begin{pmatrix} M_0 + \frac{2\mu_+}{l_B^2}(n+\frac{1}{2}) - \frac{g_e\mu_B}{2}B & \frac{\sqrt{2}\beta}{l_B}\sqrt{n} & -i\frac{\sqrt{2}\alpha}{l_B}\sqrt{n+1} & 0 \\ \frac{\sqrt{2}\beta}{l_B}\sqrt{n} & -M_0 - \frac{2\mu_-}{l_B^2}(n-\frac{1}{2}) - \frac{g_h\mu_B}{2}B & 0 & 0 \\ i\frac{\sqrt{2}\alpha}{l_B}\sqrt{n+1} & 0 & M_0 + \frac{2\mu_+}{l_B^2}(n+\frac{3}{2}) + \frac{g_e\mu_B}{2}B & -\frac{\sqrt{2}\beta}{l_B}\sqrt{n+2} \\ 0 & 0 & -\frac{\sqrt{2}\beta}{l_B}\sqrt{n+2} & -M_0 - \frac{2\mu_-}{l_B^2}(n+\frac{5}{2}) + \frac{g_h\mu_B}{2}B \end{pmatrix}.$$

(Note that the full matrix, coupling all four bands, applies only for $n > 0$; it reduces to $3 + n$ coupled bands for $n = 0$, -1, -2 since the entries' ansatz must have an oscillator index $n \geq 0$.) In the inset of Fig. 1, we show the evolution of the LL branches for a field of 0.6 T. For an increasing LL index i (labeling the states in each band), the lower branches first disperse upward, before they turn downward after reaching the hybridized gapped region around $i = 7$. When increasing the magnetic field, the distance between the levels increases such that each of the levels will be pushed consecutively over the maximum of the LL branch —a prerequisite for the AdHvAE.

With the (numerically) calculated energy levels E_n^α (with α labeling the energies of each matrix H_n), one can directly calculate the magnetization from the grand potential via

$$M = -\frac{\partial}{\partial B}\Omega, \quad \text{with} \quad \Omega = -k_B T N_\Phi \sum_{n,\alpha} \ln\left(1 + e^{(\mu - E_n^\alpha)/k_B T}\right), \tag{2}$$

with the Landau level degeneracy $N_\Phi = BA/\Phi_0$ (here, A is the area of the 2D system and $\Phi_0 = hc/e$ is the flux quantum). However, since the energies of the two lower LL branches are unbounded from below for an increasing n, the sum is divergent at its upper limit, $n \to \infty$. This is, of course, unphysical and an artifact of the continuum approximation. The principle divergence can be removed by subtracting the grand potential for a simple band insulator, obtained for noninverted levels with $M_0 \to \infty$, for which the only occupied levels are $\tilde{E}_n^1 = -(2\mu_-/l_B^2)(n + \frac{5}{2})$ and $\tilde{E}_n^2 = -(2\mu_-/l_B^2)(n - \frac{1}{2})$. This simple band insulator has a net magnetization that identically vanishes. Therefore, we can obtain the magnetization M for our model from the difference

$$\frac{\tilde{\Omega}}{k_B T N_\Phi} = -\sum_{n,\alpha} \ln(1 + e^{(\mu - E_n^\alpha)/k_B T}) + \sum_{n,\alpha=1,2} \ln(1 + e^{(\mu - \tilde{E}_n^\alpha)/k_B T}).$$

This expression does retain a weak (logarithmic) dependence on the upper energy cutoff, which provides a small smooth background to the oscillations. We show only the oscillatory component of M, which is independent of our regularization scheme, by subtracting this cutoff dependent constant [32].

A simple but experimentally crucial observation is the following: Only thermodynamic observables, e.g., the magnetization or its susceptibility, oscillate as a function of the field because they are determined by the sum over all occupied energies [8]. However, other observables like charge transport or NMR relaxation rates do not oscillate because they are determined by the DOS at the chemical potential, which is, of course, zero in the insulating regime [14]. These simply have a vanishing contribution for temperature scales below the activation gap [9].

Because the calculation of the charge conductivity requires a precise knowledge of scattering mechanisms from defects and interactions, here, we calculate only a proxy of it. The main qualitative behavior of transport properties is captured by the evolution of the temperature dependent density of states at the Fermi level (μDOS) [9],

$$\mu\text{DOS} = \sum_{n,\alpha} \frac{\partial n_F(E_n^\alpha)}{\partial \mu} = \frac{1}{2k_B T} \sum_{n,\alpha} \frac{1}{\cosh(\frac{E_n^\alpha - \mu}{k_B T}) + 1},$$

with the standard Fermi function n_F.

Results.—In the following we focus on the small-gap insulating regime of InAs/GaSb QWs, which is expected to be quantitatively described by the band structure shown in Fig. 1. The amplitude of the AdHvAE is observable only in the regime where the hybridization gap, Δ, is not considerably larger than the energy scale associated with the LL quantization of the energies [8]; otherwise, it is exponentially suppressed. This scale is given by the cyclotron frequency $\hbar\omega_c = 2\mu_+/l_B^2$ and determines the magnetic field range in which the AdHvAE is observable. For InAs/GaSb QWs, it varies from about 3 meV for $B =$ 1.7 T to 1 meV at $B = 0.6$ T, which is easily reachable experimentally.

We use the same parameters as before and show in Fig. 2 the magnetization (the red solid curve) as a function of the inverse magnetic field for a temperature $T = 0.3$ K. The inset shows the evolution of the extremal energy levels closest to the chemical potential, highlighting the fact that, for inverse fields below $1/B \approx 0.57/\text{T}$ (the black arrow), the gap closes and the system becomes metallic [9]. At this point the amplitude of the magnetization increases.

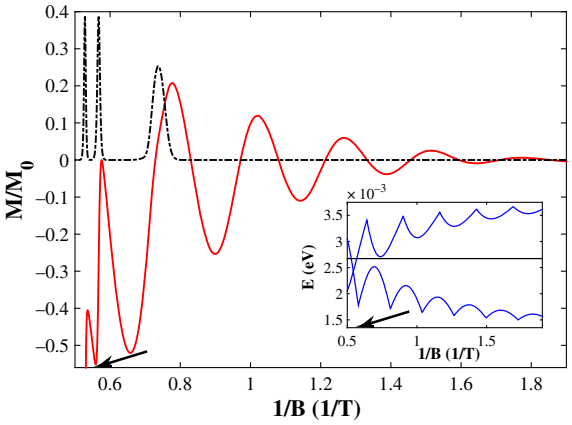

FIG. 2. QO as a function of inverse magnetic field in the narrow gap insulating regime with the same parameters as were used in Fig. 1 and for $T = 0.3$ K. The amplitude of the magnetization M (the red solid curve) is scaled by the QO amplitude M_0 in the unhybridized electronlike metallic subsystem; see the text. The evolution of the μDOS, which is expected to follow the qualitative behavior of charge transport, is also shown (the black dashed line). Note the magnetic field induced gap closing (the black arrow), which can also be traced by the evolution of the extremal energy levels around the chemical potential shown in the inset.

However, the central result of this work is that clear QOs of the magnetization persist even in the insulating regime [8]. The frequency is given by the area of the gapped shadow FS. Note that the amplitude of the oscillating magnetization M is expressed in units of the typical amplitude $M_0 = \hbar\omega_c \rho A/\pi B$ [6] for oscillations of the electronlike band in the unhybridized metallic regime (here, $\rho = A_k/2\pi^2$ is the density set by the relative FS area A_k). The amplitude of the QOs in the insulator is about an order of magnitude smaller than in the metal, but it should still be detectable in experiments.

In addition, we show in Fig. 2 the evolution of the μDOS (the black dashed line), which mimics the behavior of charge transport. Note that we have checked that, for chemical potentials inside the bands, the QO period directly scales with the FS area, as expected for the standard SdHE and dHvAE. Here, sharp peaks appear once the system becomes metallic below $1/B \approx 0.57/\mathrm{T}$ (the black arrow), but, in contrast to the magnetization, there are no oscillations in the insulating regime. The smaller broad peak appearing around $1/B \approx 0.73/\mathrm{T}$ is related to the small gap (see the inset) on the order of $k_B T$, which leads to thermally excited states at the Fermi level but which also disappears for lower T's.

In the upper panel of Fig. 3, we show the temperature evolution of M/M_0. Note the small drift as a function of temperature and field, which is related to the fact that two bands with different gap values contribute. Moreover, the gap values are dependent on the magnetic field and the complex behavior of the LL dispersion can even result in temperature induced phase jumps of the oscillations [9].

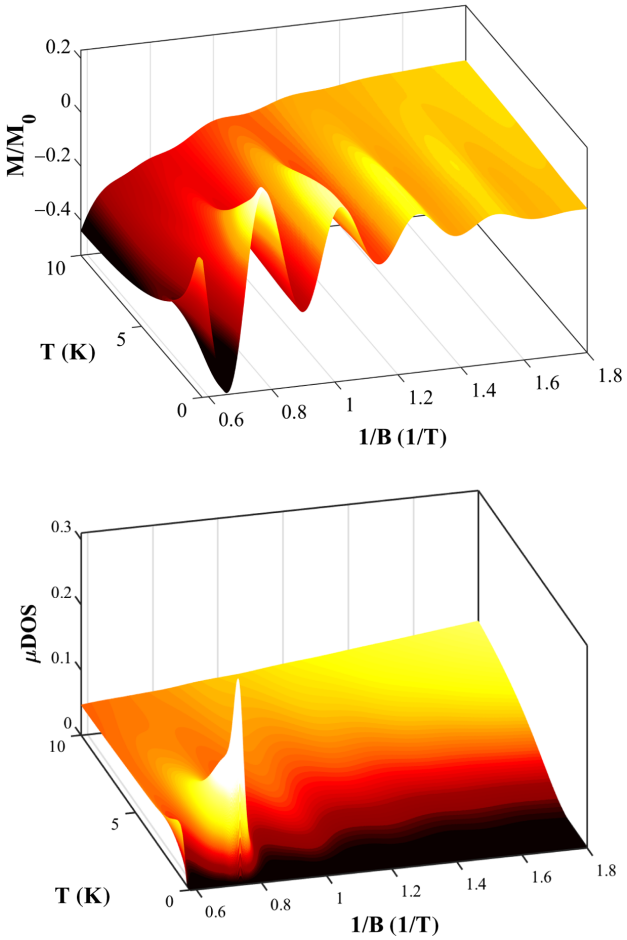

FIG. 3. The evolution of the magnetization (upper panel) and the μDOS (lower panel), which mimics the behavior of charge transport, as a function of temperature.

Both the amplitude of M and μDOS deviate substantially from the standard LK behavior. In standard metallic systems, the oscillation amplitude is always monotonically decreasing as a function of T [6], whereas in the AdHvAE it has a plateau at the lowest temperatures, or even a maximum at a temperature T^*, which is set by the distance of the LL extrema to the chemical potential [8]. In the future this could be used to measure the size of the insulating gap Δ and the position of μ. In the upper panel of Fig. 3, the maximum of the amplitude moves to higher temperatures for increasing $1/B$, which traces the evolution of the gap (see the inset) in Fig. 2. Further increasing the temperature, the amplitudes decrease until they are completely washed out around 10 K (above $k_B T \approx \hbar\omega_c$).

The lower panel of Fig. 3 displays the evolution of the μDOS. While there are no oscillations at $T = 0$ in the insulating regime, they appear at nonzero temperatures from thermally excited states. Nevertheless, they are very weak, except for the peak at $1/B \approx 0.73/\mathrm{T}$ associated with a very small gap; see the inset in Fig. 2. The weak oscillations quickly disappear with an increasing temperature.

Finally, we have checked that the behavior is qualitatively similar for different values of the Zeeman coupling (g factors) or gap values, with only slight variations arising from change of the LL gap and the critical field above which the system becomes gapless. For example, for a much bigger gap of $\Delta = 6.3$ meV ($\beta = 0.36$), we find that there are still appreciable QOs in M for fields above 3 T (not shown). Overall, the AdHvAE is quite robust, as long as the lower LL branches disperse up- and downward and Δ does not greatly exceed $\hbar\omega_c$. Hence, also, the inclusion of the BIA terms or other small perturbations will induce only small quantitative changes.

Discussion.—We have shown that the AdHvAE is observable in the narrow gap insulating regime of InAs/GaSb QWs. For reasonable band gaps $\approx$1.5 meV, magnetic oscillations appear at temperatures below about 5 K for magnetic fields around 1 T, with an amplitude that is roughly one order of magnitude smaller than for the corresponding gapless electron subsystem. Hence, the experimental constraints are well within reach, and the remaining experimental challenge should be the measurement of the magnetization of InAs/GaSb QWs, e.g., via magnetic torque, as has been carried out for other 2D electron gases [33–35]. The predicted behavior contrasts strongly with standard QOs in metals, where both the SdHE and the dHvAE occur simultaneously. For the AdHvAE in insulators, only observables related to the thermodynamic potential oscillate, whereas those quantities determined by the DOS at the chemical potential, e.g., charge transport, vanish in the low temperature limit. Hence, the observation of QOs in the magnetization without a SdHE will be an unambiguous signature of the AdHvAE. We note that the amplitude of QOs depends strongly on the hybridization gap and on the chemical potential (at nonzero T). One complication potentially arises from local variations, which can lead to gapless charge puddles [36–38]. However, the AdHvAE can still dominate over most of the temperature regime: Standard QOs from any gapless regions decay once T exceeds the (small) cyclotron energy of the hybridized bands, while the AdHvAE survives up to the (large) cyclotron energy of the unhybridized bands; see the Supplemental Material [29].

The AdHvAE effect should be observable in other (topological) insulators provided that they have small effective masses, such that the cyclotron frequency is on the order of the insulating gap and the maxima of the valence band traces out a well-defined area in momentum space. The latter condition disqualifies, for example, HgTe QWs, but other systems, e.g., gated bilayer graphene, fall into that category. In the future, we hope the AdHvAE can be turned into a powerful tool for measuring gap sizes and the position of the chemical potential in this class of narrow gap insulators.

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