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Deep-lying semi-Dirac fermions in hexagonal close-packed cadmium

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Semi-Dirac fermions are massless in one direction and massive in the perpendicular directions. Such quasiparticles have been proposed in various contexts in condensed matter. Using first principles calculations, we identify a pair of semi-Dirac bands anti-crossing at -3 eV below the Fermi level in the electronic structure of hexagonal close-packed (hcp) cadmium. The linear out-of-plane dispersion is kept up to the Fermi level. We demonstrate that the dichotomy between the linear and quadratic dispersions is driven by an orientation-sensitive hybridization between the s and p_z orbitals. The upper semi-Dirac band produces a lens-shaped nonellipsoidal Fermi sheet whose cross-section area has a k -dependence that is in excellent agreement with the experimentally measured period of Sondheimer oscillations.

I. INTRODUCTION

Linearly dispersive electronic bands can give rise to massless analogues of relativistic particles in condensed matter systems. Such Dirac fermions lead to intriguing experimental consequences like the $E_N \propto \sqrt{NB}$ energy spectrum of the Landau levels, half-integer anomalous quantum Hall effect, and Klein tunneling [1]. Observation of these phenomena in monolayer graphene [2], which has Dirac cones at its Brillouin zone corners, has motivated the search for other such systems.

Semi-Dirac fermions exhibiting direction-dependent massless and massive behaviors have attracted much attention during the last two decades. They were proposed to exist in the A phase of ^3He [3], in a graphenelike model subjected to a magnetic field [4], VO_2 layers confined within TiO_2 [5], strained silicene oxide [6], and tetragonal perovskite oxides with the $I4/mcm$ space group [7]. The merging-of-Dirac-points scenario provides a unifying topological picture for this phenomenon [8, 9], with Bloch-Zener dynamics across the transition analyzed in [10]. A magnetic-flux analogue appears in the Hofstadter spectrum [11], and transport and criticality near the merging transition were analyzed in Refs. [12, 13]. While their observations have been reported in microwave analogue of a graphenelike lattice [14], honeycomb lattice of micropillars [15], and cold atoms [16], they have been more elusive in solids. Recently, a semi-Dirac dispersion leading to a $[(N + \frac{1}{2})B]^{2/3}$ scaling of the Landau levels [4, 17] has been reported in ZrSiS [18].

Generally, linear dispersion is expected when two non-bonding bands cross at a Dirac point situated away from the Brillouin zone center. Semi-Dirac bands in $(\text{TiO}_2)_5/(\text{VO}_2)_3$ arise in this way [5]. Another avenue is the inversion of the relative positions of bands associated with s and p orbitals. In HgTe-CdTe quantum wells, such an inversion occurs at a critical thickness leading to a Dirac point at the Brillouin zone center [19]. Given that s - p hybridization is uniform along all three axes in

cubic zinc blende HgTe , however, a semi-Dirac analogue of this mechanism has been missing.

A possible avenue to obtain semi-Dirac bands is to explore cases with direction-dependent s - p hybridization. Metals in column twelve of the periodic table are promising, as their mobile electrons originate from s and p orbitals rather than d orbitals. Furthermore, unlike column eleven elements (the noble metals) with a nominal electronic configuration s^1 , column twelve metals are nominally s^2 . The additional electron in column 12 metals favors the hcp structure, where the p_z orbital hybridizes differently with the s orbital compared to the p_x and p_y orbitals.

In this paper, we use first principles calculations to show that hcp Cd hosts a pair of semi-Dirac bands anti-crossing at -3 eV below the Fermi level. The bands' dispersion is parabolic in plane and linear out of the plane over an extended distance in the reciprocal space that includes the Brillouin zone center and the Fermi level crossing. The two bands have mixed s and p_z character in the out-of-plane direction and unmixed s or p_z character in the in-plane directions. Thus, the direction-dependent mixing of the s and p_z characters is responsible for the presence of the semi-Dirac bands. Comparison with hypothetical hcp Ag shows that the relative positions of the s - and p_z -like bands control the direction-dependent mixing. We show that in such a hypothetical hcp metal with a single s electron, there is a large gap between s -like and p_z -like states. Consequently, s - p_z hybridization is negligible and the bands are parabolic in all directions. We find that tensile strain moves the s -like states higher relative to the p_z states, thereby increasing the s - p_z hybridization in the out-of-plane direction. Semi-Dirac dispersion occurs near the point of s - p_z inversion, in a manner analogous to that of HgTe-CdTe quantum wells where the band inversion is controlled by tuning the strength of the effective spin-orbit interaction. The Fermi level crossing of the upper semi-Dirac band generates a non-ellipsoid Fermi pocket, called lens-shaped in the scientific litera-

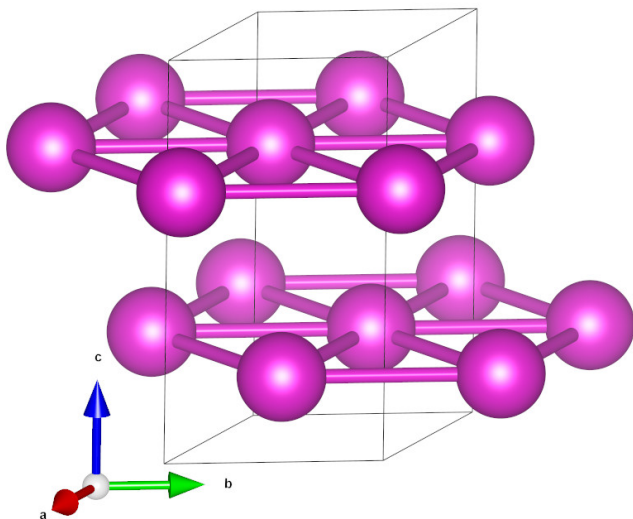


FIG. 1. Hexagonal closed packed structure of cadmium. The inter- and intra-layer nearest-neighbor Cd-Cd distances are 3.293 and 2.979 Å, respectively.

ture. We find that the calculated derivative of the cross-section area of this sheet with respect to the out-of-plane wave vector agrees with the experimental value extracted from the observed Sondheimer oscillations, corroborating the presence of the calculated semi-Dirac bands in this material.

II. COMPUTATIONAL DETAILS

The first principles calculations presented here were performed using the generalized full-potential method as implemented in the WIEN2K package [20]. We used the local density approximation (LDA) for the exchange-correlation functional and muffin-tin radius R of 2.5 a.u. The plane-wave cutoff K_{\max} was set using the condition $RK_{\max} = 7$. A $72 \times 72 \times 36$ k -point grid was used for the Brillouin zone integration. The SKEAF code was modified inhouse to output the cross-section area of the Fermi sheet as a function of the k_z wave vector [21]. The effective mass was obtained by fitting the semi-Dirac band along the inplane direction by the polynomial $E(k) = E_0 + \hbar^2 m k^2 / 2$ around $\pm 0.05 \text{ \AA}^{-1}$ near the Fermi level crossing. Results presented in the paper do not include the spin-orbit interaction. However, we verified that inclusion of spin-orbit interaction does not change the dispersion of the semi-Dirac bands.

The hexagonal close packed structure of cadmium is shown in Fig. 1. We used the experimental lattice parameters $a = 2.979$ and $c = 5.617 \text{ \AA}$ in all the calculations [22]. The two atoms in the unit cell are situated at $(1/2, 2/3, 1/4)$ and $(2/3, 1/3, 3/4)$. Note that the c/a ratio of 1.886 of hcp Cd is much larger than the ideal value of $\sqrt{8/3} \sim 1.633$ required for the densest packing. The deviation likely results from the additional complexity of

interatomic bonding due the presence of the $4d$ states near the Fermi level. Contrast this to hcp Mg (that does not have $3d$ states), where $c/a = 1.623$.

III. RESULTS AND DISCUSSION

The calculated LDA band structure of hcp Cd is shown in Fig. 2. There are ten narrow bands with $4d$ character between -10 and -8 eV relative to the Fermi level. Dispersive bands with $5s$ and $5p$ characters lie above and cross the Fermi level. The calculated Fermi surface of hcp Cd is shown in Fig. 2b-d. It consists of two hole-type sheets known as “caps” and “monster” centered around the six edges of the Brillouin zone and the electron-type lens sheet centered at Γ . The volume enclosed by these sheets are 0.01 and 0.19 holes and 0.20 electrons per cell, respectively. The corresponding carrier concentrations are 0.17 , 3.31 and $-3.48 \times 10^{22} \text{ cm}^{-3}$.

Our results generally agree with those obtained by Stark and Falicov using empirical pseudopotential method [23] and Daniuk et al. through self-consistent first principles method [24], although there are differences. All three results show one band crossing the Fermi level along ΓA and ΓM , while two bands cross along ΓK , HA , and HL . One band in our and Stark and Falicov’s calculations traverses the K point at a saddle point, whereas such a crossing is missing in that of Daniuk et al. Furthermore, the doubly degenerate band that almost touches the Fermi level at the L point in our calculations lie noticeably higher in the other two results.

A prominent feature of the electronic structure of hcp Cd is the pair of bands that linearly disperse away from -3 eV in opposite directions along the out-of-plane path ΓA . In Stark and Falicov’s study, these bands appear to taper off quadratically when they approach each other at Γ . However, the linear dispersion, although not explicitly noted, is visible in the results of Daniuk et al. [24]. Interestingly, these bands disperse quadratically along ΓK and ΓM . This implies that the electronic states are massless in the out-of-plane direction and massive within the plane. We obtain $0.8m_e$ for the inplane effective mass by a quadratic fit near the Fermi level crossing. It is also striking that the hole-type linearly dispersing band along out-of-plane direction becomes electron-like when it shows quadratic dispersion along the in-plane directions. These pair of bands should accurately be called $1/3$ -Dirac bands. However, we follow the convention in the literature and call them semi-Dirac bands.

Semi-Dirac points previously discussed in the literature are associated with degeneracies due to band crossings [4, 5, 11, 17]. In the present case, there is a gap of 50 meV at Γ near -3 eV where two semi-Dirac bands converge even without spin-orbit interaction. To disentangle the role of structure and chemistry behind the origin of these semi-Dirac bands, let us perform a gedanken experiment by supposing that the next-door element in the periodic table Ag, instead of being face-centered cubic (its sta-

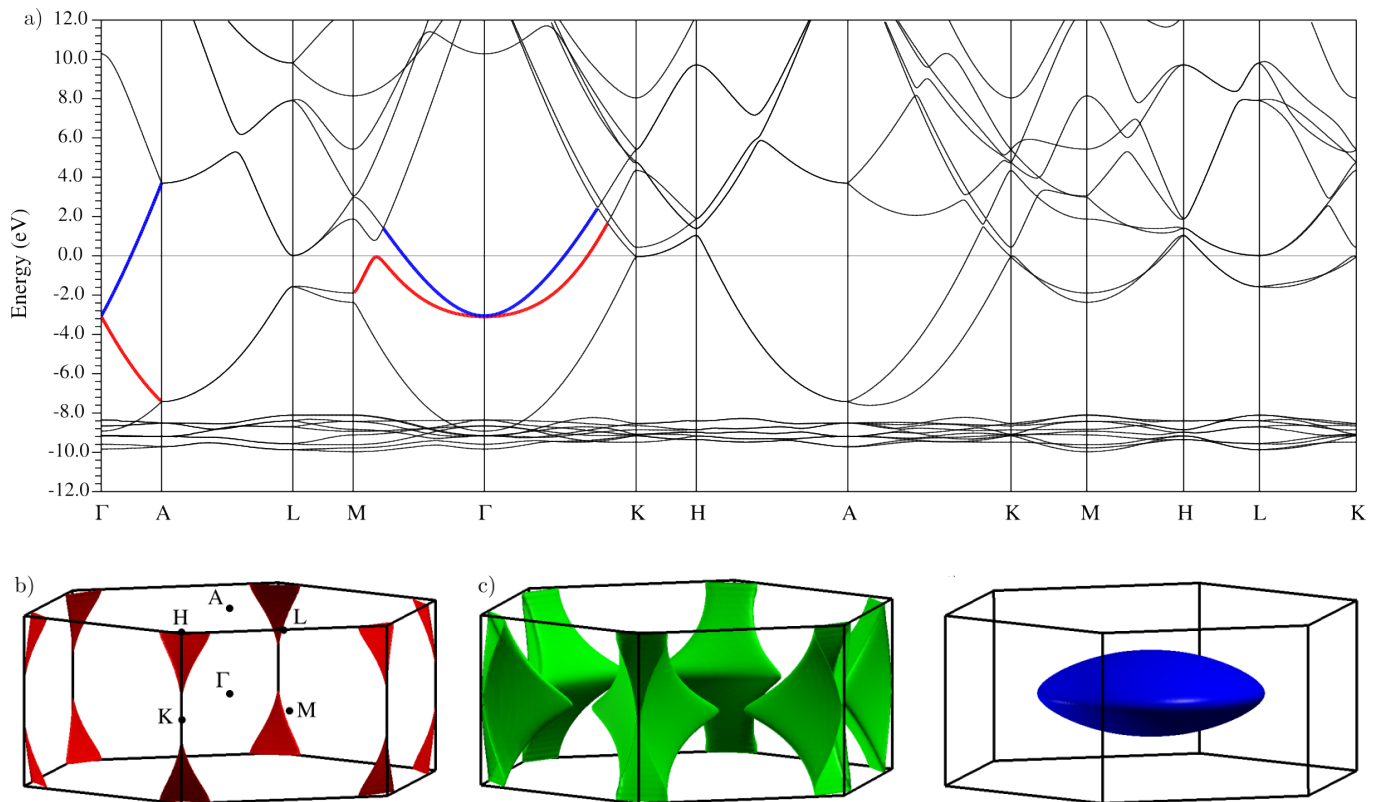


FIG. 2. a) Calculated LDA band structure of hcp Cd. The band structure features a pair of bands that disperse linearly along the out-of-plane direction ΓA and quadratically along the inplane directions ΓM and ΓK . These bands appear to converge near -3 eV at Γ , but they are in fact separated by 50 meV. The upper and lower semi-Dirac bands are shown in blue and red, respectively. Calculated Fermi surface of hcp Cd consisting of b) hole-type “cap”, c) hole-type “monster”, and d) electron-type “lens” sheets. The lens sheet derives from the upper semi-Dirac band.

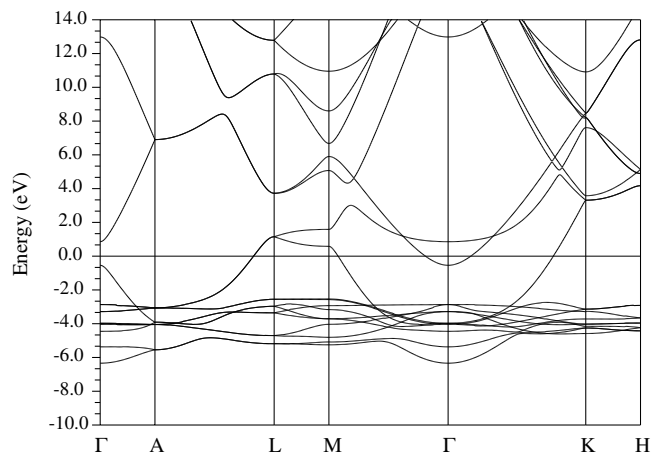


FIG. 3. Calculated LDA band structure of a *hypothetical* hcp Ag using the lattice parameters of hcp Cd. The pair of bands that linearly disperse along ΓA in hcp Cd disperse quadratically in hypothetical hcp Ag. The gap at Γ between these bands also increases to 1.4 eV.

ble structure), could be synthesized in the hcp structure of Cd. Fig. 3 shows the band structure of such a hypo-

thetical hcp Ag with the same structural parameters as that of hcp Cd. We can see that the band structures of isostructural hcp Ag and Cd differ by more than a rigid shift of the Fermi level even though they lie next to each other in the periodic table and their electron count differs only by one. Hcp Ag no longer exhibits the linearly dispersing bands along the out-of-plane direction ΓA . The gap between these bands increases to 1.4 eV, and they taper off parabolically as they approach Γ . Therefore, a crossing of nonbonding bands do not seem to play a role in the appearance of the semi-Dirac bands in hcp Cd. Furthermore, the contrasting electronic structures of isostructural hcp Cd and Ag shows that the semi-Dirac bands do not arise merely due to the layered hexagonal structure and points out the essential role played by the chemistry of divalent $5s^2$ electronic configuration in Cd.

We have plotted the band structures of hcp Cd and Ag showing the contribution of s and p_z orbital characters along the out-of-plane and two in-plane directions in Fig. 4 to understand the changes in band hybridization due to the presence of an additional $5s$ electron in Cd. Along the out-of-plane ΓA direction, we can see that the pair of semi-Dirac bands in hcp Cd show nearly equal amount of s and p_z characters, especially near Γ . In hcp

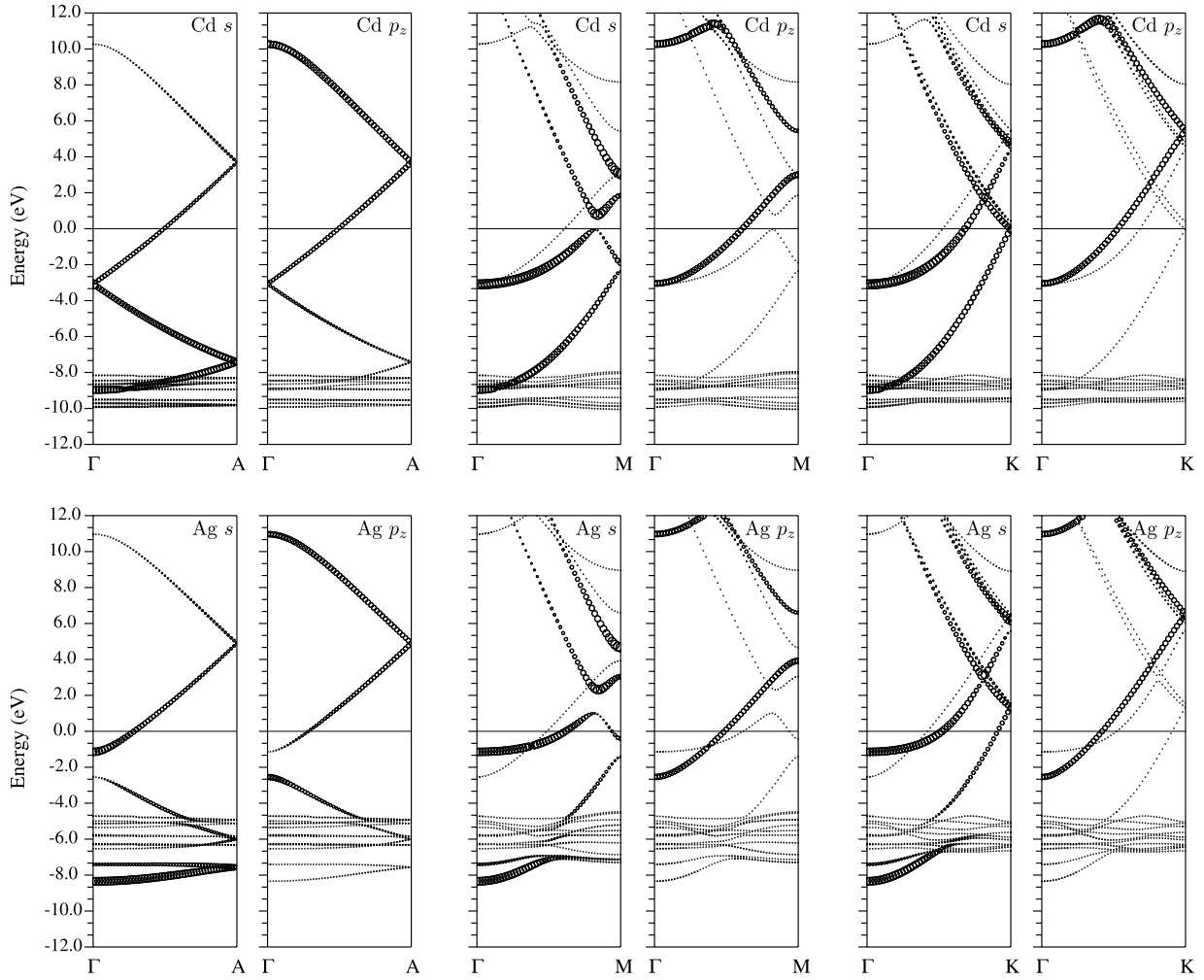


FIG. 4. Band structures of hcp Cd and hypothetical hcp Ag isostructural to hcp Cd as a function of respective s and p_z orbital characters along out-of-plane ΓA and inplane ΓM and ΓK directions. The semi-Dirac bands in hcp Cd show a mixed s and p_z character in the out-of-plane direction. In hcp Ag, the bands disperse quadratically in all directions, and the upper band is predominantly s -like, while the lower band is mainly p_z -like.

Ag, in contrast, the upper-lying band shows dominant s character, while the p_z character is dominant in the lower-lying band. This is particularly the case close to Γ , where the upper-lying and lower-lying bands are almost solely due to the s and p_z states, respectively. Thus, s - p_z hybridization along one orientation gives rise to a linear dispersion, and the lack of it along the two others leads to a quadratic one. This observation is further reinforced looking at the orbital characters of these bands along ΓM and ΓK . In both materials, along these in-plane directions, each band exhibits one dominant orbital character, and all of them disperse quadratically.

Interestingly, the higher-lying band along the in-plane directions has p_z character in hcp Cd but s character in hcp Ag. This band inversion is reminiscent of the one noted in HgTe-CdTe quantum wells [19]. When the well is thin, the s -derived band lies higher than the p -derived band, the orbital mixing is low, and the bands are quadratic with a finite gap at Γ . With increasing thick-

ness of HgTe in the quantum well, the orbital characters start to mix, the gap decreases, and the bands start to become linearly dispersive. The gap vanishes and the bands cross each other linearly at a critical thickness, beyond which the higher-lying band exhibits more p character. In this analogy, hcp Ag is like thin HgTe below the critical thickness and its s -type band lies above its p_z -type band. Meanwhile, hcp Cd is analogous to thick HgTe above the critical thickness, because its p_z -type band lies just above its s -type band.

Since the band structures of hcp Cd and hcp Ag have been calculated using the same lattice parameters, the difference in the relative position of their s - and p_z -type bands near the Fermi level is due to the presence of an additional electron in Cd. The extra electron does not just rigidly fill up the unoccupied states in the band structure when going from hcp Ag to hcp Cd, it also pulls down the s -type states relative to the position of the p_z -type states. We discuss the inversion of the relative positions

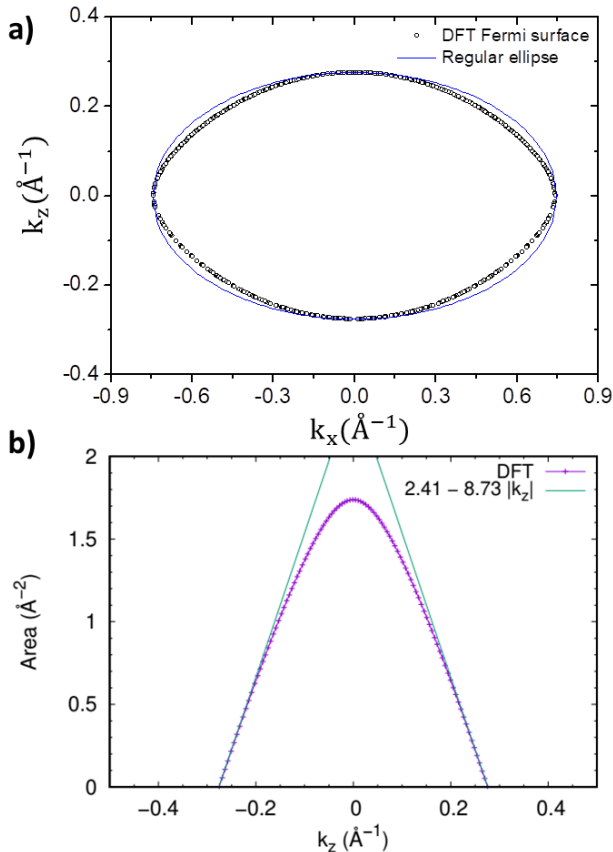


FIG. 5. a) The electron-like Fermi surface generated by the upper semi-Dirac band projected in the k_x, k_z plane (empty circles). The blue solid line represents a regular ellipsoid whose axes are equally long. Note the difference. b) The cross section area as a function of k_z . Over a long distance, near the poles (that is $k_{z,min}$ and $k_{z,max}$), the area is linear in k_z , consistent with the linear dispersion of the semi-Dirac band from which this Fermi sheet derives.

of the s - and p_z -type states in hcp Ag due to tensile strain in the appendix.

Direct observation of the linear dispersion of these semi-Dirac bands in hcp Cd emerges as a task for future experiments employing angle-resolved photoemission spectroscopy. However, it has already a striking experimental signature in the Sondheimer oscillations [25] of cadmium detected by experiments [26, 27].

In a metallic crystal hosting ballistic electrons, galvanometric coefficients display oscillations, which are periodic in magnetic field. The period of these oscillations is given by [28, 29]:

$$\Delta B = \frac{\hbar}{ed} \left(\frac{\partial A}{\partial k_z} \right), \quad (1)$$

Here, d is the thickness of the crystal in the direction perpendicular to the magnetic field, e is the electron charge, \hbar is the reduced Planck constant, k_z is the component of the wave vector along the magnetic field, and

A is the cross section area of the Fermi sheet intersecting with a plane perpendicular to k_z .

In a perfect ellipsoid, $\frac{\partial A}{\partial k_z}$ is not constant and smoothly varies as function of k_z . At the limit point (the apex) of the ellipsoid, it has a slightly higher degeneracy than everywhere else. With increasing number of states sharing an identical $\frac{\partial A}{\partial k_z}$, oscillations sharing the same periodicity become more robust. In our case, the electron-like pocket formed by the upper semi-Dirac band (Fig. 2d) is not an ellipsoid. Fig. 5a shows how the projection of this Fermi surface in the (k_x, k_z) plane is not a regular ellipsoid. Fig. 5b shows its cross section area as a function of k_z . Along this orientation, the apices of this lens-like Fermi surface occur at $\pm 0.28 \text{ \AA}^{-1}$. The area is proportional to k_z along a large fraction of the out-of-plane path in the Brillouin zone near these points, as expected by the linear dispersion in this direction. The figure also shows a linear fit, which demonstrates that the quadratic component is negligible up to a distance of 0.09 \AA^{-1} towards the Brillouin zone center from the apices. The slope of the linear fit is $\frac{\partial A}{\partial k_z} = 8.73 \text{ \AA}^{-1}$.

This calculated slope is in excellent agreement with what was found experimentally first by Grenier et al. [26] in a sample with $d = 1.02 \text{ mm}$. According to their data, $\delta B = 0.0564 \pm 0.0003 \text{ T}$. Inserting these in Eq. 1, yields $\frac{\partial A}{\partial k_z} = 8.76 \text{ \AA}^{-1}$. A recent experiment [27] quantifying the periodicity across a forty-fold variation of thickness found an identical value of 8.76 \AA^{-1} . Now, if the deviation from the ellipsoidal shape (see Fig. 5a) is neglected, the expected period is the apex $\frac{\partial A}{\partial k_z} \Big|_{F_z} = 2\pi \frac{k_{F_r}^2}{k_{F_z}}$, which yields 12.58 \AA^{-1} . Such a large 44% difference confirms the importance of the linear dispersion along k_z in setting the period of the Sondheimer oscillations observed in this material. The agreement between theory and experiment implies that the slope of the out-of-plane linear dispersion near the Fermi level conforms to what is computed here.

IV. CONCLUSIONS

In summary, revisiting the electronic structure of hcp Cd using first principles calculations demonstrates that it hosts deep-lying anti-crossing of semi-Dirac bands with linear out-of-plane dispersion and quadratic in-plane dispersion. We argue that this is driven by direction-dependent mixing of s and p_z states, and that hcp Cd can be likened to HgTe-CdTe near its critical thickness, except with the restriction of band hybridization along a single direction. The nonellipsoid electron pocket produced by the upper semi-Dirac band has a cross-section area proportional to the out-of-plane wave vector along a significant fraction of the Brillouin zone path and is in excellent quantitative agreement with the period of Sondheimer oscillations.

V. ACKNOWLEDGEMENTS

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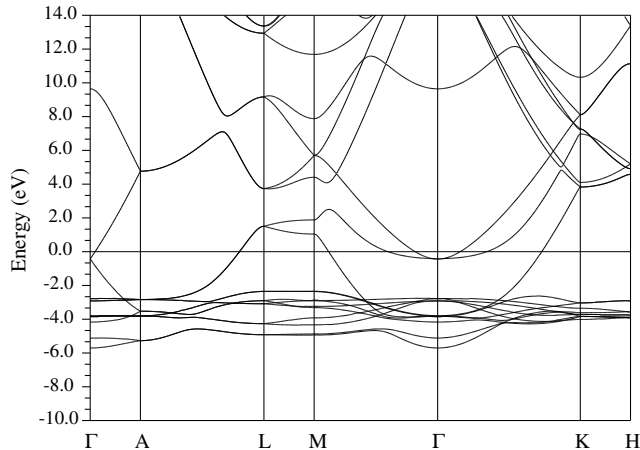


FIG. 6. Calculated LDA band structure of hypothetical hcp Ag with the lattice parameter c strained by 13% compared to that of hcp Cd. Semi-Dirac bands appear in hcp Ag when strained tensilely.

Appendix: Engineering band dispersion with strain

The relative position of the s - and p_z -type bands near the Fermi level in hcp Ag and the hybridization between them can be controlled by varying the lattice parameter c . As c is increased, the gap at Γ between the s and p_z states decreases, and these bands become more linear near Γ along the out-of-plane direction. The gap vanishes when c is increased by 13%, as can be seen in Fig. 6 that shows the band structure of hcp Ag at this value of the lattice parameter. We can also see that these bands converge at a semi-Dirac point, with linear dispersion along the out-of-plane direction Γ - A and parabolic dispersion along the in-plane directions Γ - M and Γ - K near the Brillouin zone center.

Fig. 7 shows the corresponding band structure weighted with the s and p_z orbital contributions along the out-of-plane and two inplane paths, and they are now similar to that of hcp Cd. Along the out-of-plane direction, the two semi-Dirac bands show almost equal amount of s and p_z contribution. There is very little

hybridization between these states in the inplane directions, with the p_z -type band lying above the s -type band away from Γ as in the case of hcp Cd. This confirms that direction-dependent band hybridization driven by s - p_z inversion forms the semi-Dirac bands in these systems. In the case of HgTe-CdTe quantum wells, the band inversion is caused by spin-orbit interaction. Here, the inversion is tuned by interplanar distance. Of course, the

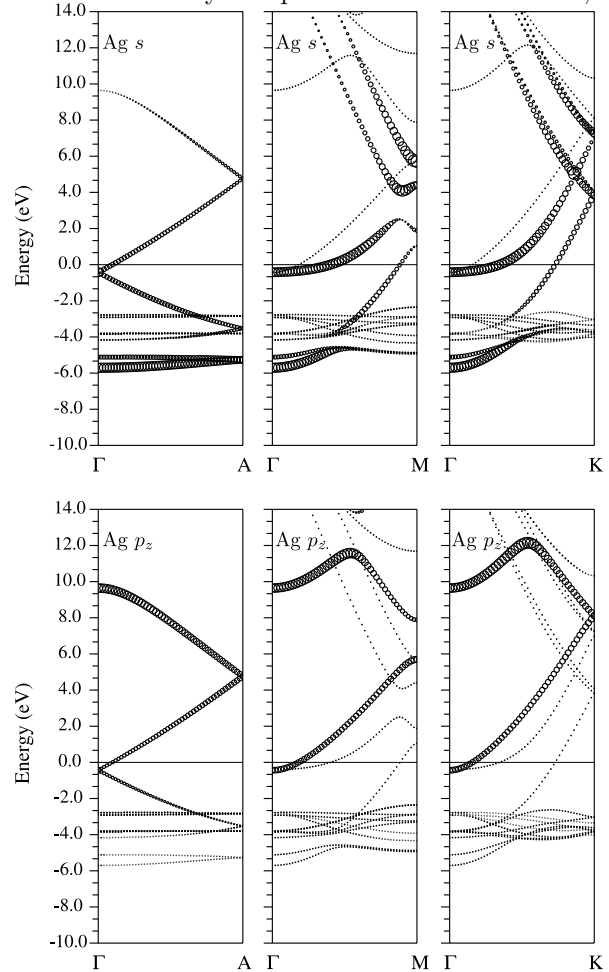


FIG. 7. Same as in Fig. 6, but plotted as a function of s and p_z orbital characters along the out-of-plane ΓA and in-plane ΓM and ΓK directions. The linearly dispersive bands in the out-of-plane direction show mixed s and p_z characters, while the bands remain unmixed in the inplane directions. The unmixed s -like band lie below the unmixed p_z -like band, indicating that tensile strain inverts the relative positions of these bands.

strain value of 13% is highly unphysical and this hypothetical scenario mainly serves to provide an example of band inversion due to strain.

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