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Dirac-Like Electronic-Band Dispersion of $LaSb_2$ Superconductor and Its Counterpart $LaAgSb_2$

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Electronic properties of the $LaSb_2$ superconductor and its non-superconducting $LaAgSb_2$ analogue are determined within the density functional theory approach. The existence of 2D Dirac fermions in the bulk electronic structure of both compounds is unveiled and analysed based on weighted bands calculated around the Fermi level. Interestingly, a lack of superconductivity in $LaAgSb_2$ can be explained in terms of the topological Feshbach shape resonance of some Fermi surface sheets, associated with the reduction of their dimensionality (from 3D to 2D).

topics: electronic structure, DFT calculations, Dirac materials, superconductivity

1. Introduction

The layered $LaSb_2$ and $LaAgSb_2$ intermetallics belong to the family of anisotropic rare-earth diantimonides and show fascinating physical properties. These compounds reveal, e.g., a charge density wave (CDW) and a significant linear magnetoresistance (MR) with no saturation in the high-field limit (up to magnetic fields of 45 T) [1]. In particular, $LaSb_2$ exhibits a pronounced difference in values of transverse and longitudinal MR (transverse MR exceeds longitudinal MR by an order of magnitude) which reflects strong anisotropy of the system [2]. However, only $LaSb_2$ shows the superconducting (SC) state below the critical temperature$T_c \approx 1.2$ K [3, 4] and complex SC phase diagrams [4], contrary to $LaAgSb_2$ that does not undergo transition into any SC state at least down to 0.5 K [5].

For both systems, we present here bulk band structures calculated from first principles, using density functional theory (DFT) methods and we discuss their densities of states (DOS), electronic bands with orbital contributions (the so-called fat bands) and the Fermi surfaces (FS). By considering mainly our calculated fat bands and taking into account that $LaAgSb_2$ was recognized earlier as a Dirac multi-band metal [6], we conclude that the linear MR in $LaSb_2$ originates also from the Dirac-like bands near the Fermi level E_F. We expect that our findings will be helpful for a better understanding of linear MR because this issue is still under debate for various materials [7, 8]. Finally, the possible explanation of the occurring SC state in $LaSb_2$ and its lack in the reference system is also discussed.

2. Computational methods

All calculations were performed with the full-potential local-orbital FPLO-14 code [9]. The Perdew–Wang parametrization of the local density approximation (LDA) was used in both the scalar and fully relativistic modes (the latter containing spin–orbit coupling (SOC)) [10, 11]. Experimental values of the lattice parameters and atomic positions of the tetragonal $LaAgSb_2$ ($ZrCuSi_2$-type structure; space group $P4/nmm$, No. 129) [12] and orthorhombic $LaSb_2$ ($SmSb_2$-type structure; space group $Cmca$, No. 64) [13] unit cells (u.c.), respectively, were assumed. These two types of u.c. are presented in Fig. 1b and 1d. Both crystal structures are centrosymmetric but non-symmorphic. The free atomic positions of La and Sb atoms were additionally optimized in the FPLO-14 code, using the Hellmann–Feynman forces. This optimization, however, only negligibly affected the electronic structures. In turn, total energy values were converged for the $16 \times 16 \times 16$ $\boldsymbol{k}$-point mesh, corresponding to 405 and 657 $\boldsymbol{k}$-points in the irreducible part of the Brillouin zone (BZ) for $LaAgSb_2$ and $LaSb_2$, respectively.

3. Results and discussion

The total DOS's plotted in Fig. 1 for $LaSb_2$ and $LaAgSb_2$ have, in general, quite a similar shape, particularly in the very vicinity of E_F. The only clear differences between them come from the prominent Ag 4d-electron contribution in the case of $LaAgSb_2$. This broad Ag 4d maximum is located mainly from -6 eV to about -3.5 eV with the inherent tail spreading up to E_F. However, in

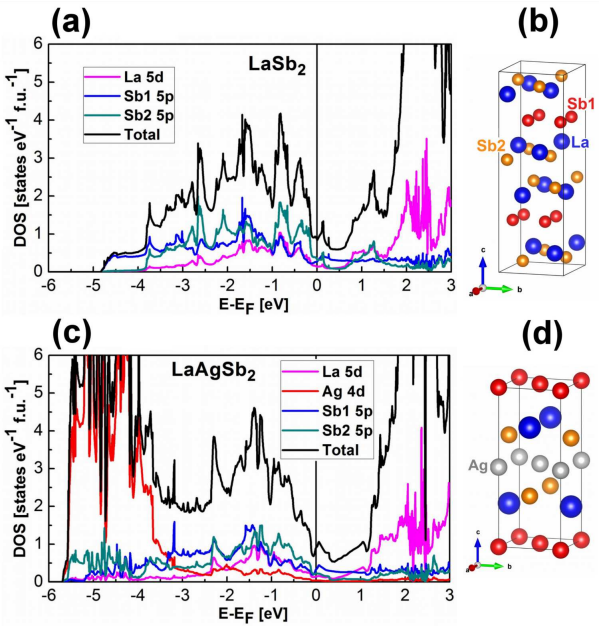

Fig. 1. The fully relativistic total and partial DOS's calculated for (a) $LaSb_2$ and (c) $LaAgSb_2$. The conventional unit cells of these compounds (shown in the right parts of the figure, (b) and (d), respectively) contain the quasi-two-dimensional (quasi-2D) nets of Sb1 atoms (called planes) that are crucial for the existence of strongly dispersive linear electronic bands near E_F (see the text and the next figure). The images of the crystal structures were drawn with VESTA [14].

this compound, even the entire Ag contribution to the total DOS near E_F (see Fig. 1b) is the smallest one in comparison with those of the remaining atoms. This feature was also observed experimentally in the photoemission spectra measured by Shi et al. [15].

The overall DOS shape for $LaSb_2$ also agrees quite well with the experimental peaks determined from the angle-resolved photoemission spectroscopy (ARPES), reported by Acatrinei et al. [13]. The values of DOS at E_F, $N(E_F)$, in these two intermetallics are very close to each other (0.835 and 0.911 states/(eV f.u.), for $LaSb_2$ and $LaAgSb_2$, respectively) and are relatively small which reflects a semimetallic character of their non-SC states. However, $N(E_F)$'s in these two compounds differ only by the Ag tail-contribution, mentioned above. Those values allow an estimation of the Sommerfeld coefficients γ from the simplest free-electron formula. Thus evaluated values of γ are equal to 1.97 and 2.15 mJ/(mol K^2) for $LaSb_2$ and $LaAgSb_2$, respectively. The latter value is in very good agreement with the experimental one, 2.62 mJ/(mol K^2), obtained for a polycrystalline sample of $LaAgSb_2$ [5]. In both cases, the total Sb $5p$ contributions prevail over the other ones. However, a more precise analysis of the Sb contributions reveals that in $LaAgSb_2$ the Sb1 $5p$ electrons yield clearly predominant inputs to $N(E_F)$, whereas in $LaSb_2$ the Sb1 $5p$ and Sb2 $5p$ electron contributions are comparable to each other at E_F. It may indicate a more 3D character of the electronic structure in $LaSb_2$ than that in the other system.

The orbital character of the electronic bands in the vicinity of E_F, displayed in Fig. 2, ensures even better insight than DOS itself into low-energy electronic properties of the systems. One can see that both in $LaSb_2$ and $LaAgSb_2$ the highly-dispersive (conic-like) linear bands, around E_F, are distinctly dominated by the $5p_{x,y}$ electron orbitals coming from the rectangular nets of the Sb1 atoms. In turn, the remaining orbital contributions are minor in this region. This situation is analogous to those of other layered 112-type intermetallics. In particular, for the $LaAgSb_2$ system it is in good accord with the results of calculations (performed by a different DFT method) and ARPES measurements, reported previously [6, 15, 16]. The explicitly linear conic-like bands are also responsible for low effective masses of the carriers in $LaSb_2$ and $LaAgSb_2$, measured in quantum oscillations experiments [17, 18]. Interestingly, values of effective masses, detected in the above experiments, are comparable for both systems investigated here ($m^* \approx 0.17 m_0$, where m_0 is the electron rest mass) [17, 18], and they are also comparable with those of another Dirac system, $LaAgBi_2$ [19]. The electronic structures are only negligibly modified by the SOC exactly at E_F, despite the distinct $5p$-electron contributions from heavy Sb atoms in this energy region. Interestingly, the optical conductivity measured for $LaSb_2$ has similar features to those of the light ZrSiS compound, i.e., the spectra are flattened and also have similar details [20, 21]. Just for the ZrSiS material, the 2D Dirac fermions in the bulk were predicted and observed experimentally [21, 22]. This may indicate good accordance with our calculation results as well as the presence of the Dirac fermions in the electronic structure near E_F, in the systems analyzed in this paper.

In the case of the Dirac fermions, unlike classical ones (with a parabolic dispersion relation), the formula for energy of the Landau levels (LL) is $E_n = \pm v_F \sqrt{2e\hbar B n}$ [19], where v_F is the Fermi velocity, B is the magnitude of external magnetic field and $n = 0, 1, 2, \ldots$. The zero-th LL in that manner is constrained only to the relativistic fermions. Note that in this case the position of the lowest Landau level (i.e., for $n = 0$) does not depend on the magnetic field, B, in contrast to the classical case, where $E_0 \sim B$. What is important, the so-called quantum limit (QL) of the Dirac fermions is considered to be crucial for observing the high-field unsaturated linear MR. The QL occurs if only the lowest Landau level (0th LL) is occupied by the carriers. It takes place only in sufficiently high magnetic fields and at low temperature. A formula for MR of the Dirac fermions in QL, obtained by Abrikosov [23, 24], reads MR $\sim (N_i/n_0^2)B$,

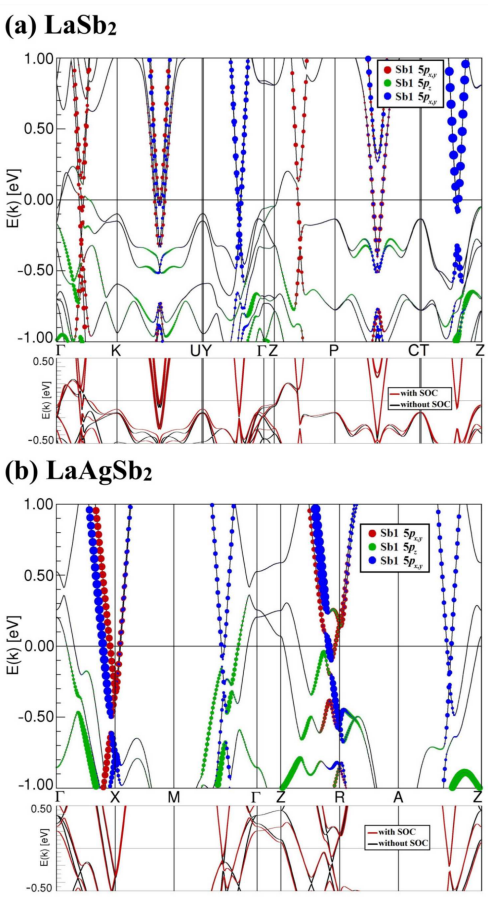

Fig. 2. The fully relativistic electronic band structures with selected orbital contributions (marked by colored circles) calculated for (a) $LaSb_2$ and (b) $LaAgSb_2$. Note that the strongly linear bands originate from the Sb1 $5p_{x,y}$ orbitals. In addition, the comparison between fully (red lines) and scalar (black lines) relativistic bands computed for each compound is attached at the bottom of a respective part. The energy gap induced by SOC is rather small (approximately of 20–50 meV).

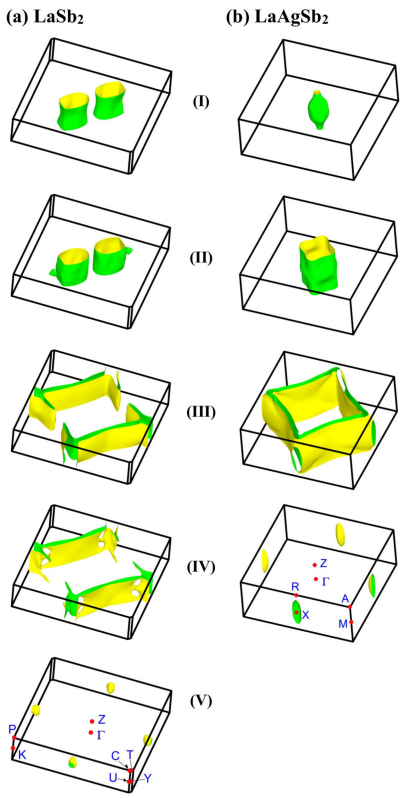

Fig. 3. The fully relativistic FS sheets of (a) $LaSb_2$ and (b) $LaAgSb_2$ compounds, originating from different conduction bands and numbered by growing Roman numerals (I–V) with increasing energy of the bands. In both systems, sheets I and II are hole-like while III–V — electron-like. The strongly quasi-2D character of some nested FS sheets (in particular III) is associated with the highly-dispersive linear electronic bands formed by the Sb1 $5p_{x,y}$ electrons.

where N_i is the concentration of the scattering centers and n_0 is the density of the Dirac carriers in the system (i.e., electrons or holes). The Hall conductivity measured for both compounds is non-zero [1, 6] which is qualitatively in good agreement with a lack of the carrier compensation ($n_e/n_h \approx 1.4$ for $LaSb_2$ and $n_e/n_h \approx 1.6$ for $LaAgSb_2$), estimated in this work based on the volumes of calculated FS sheets presented in Fig. 3. By combining that with the character of weighted bands, plotted in Fig. 2, we obtain a confirmation that the unsaturated linear MR in compounds investigated here can be explained by the presence of the Dirac-like linear bands in the vicinity of E_F.

Finally, the Fermi surface, drawn in Fig. 3a, calculated for $LaSb_2$, reveals both 3D and quasi-2D sheets. In turn, the FS sheets of $LaAgSb_2$ (in particular sheets II and III in Fig. 3b) are strongly quasi-2D and nested, resembling results of earlier calculations (using different codes) for the same system as well as those for other isostructural 112-type intermetallics, like $LaCuSb_2$ [16, 18]. It is worth underlining that the most quasi-2D electron FS sheets, numbered as III and being similar for $LaSb_2$ and $LaAgSb_2$, can be associated with the quasi-2D Dirac fermions.

Interestingly, for the latter system the hole-like FS sheets, centered at the Γ point, denoted by I and II numerals in Fig. 3b, were found by the ARPES measurements [25]. For the same system, the FS electron sheet, marked by III in Fig. 3b, was detected also by means of ARPES by Shi et al. [15]. The Fermi velocities calculated for $LaAgSb_2$ are the highest along the Γ–M line and equal to 1.3 and 1.5×10^6 m/s for two Dirac branches which is in line with the experimental values reported in [15]. In turn, for $LaSb_2$ the corresponding branches are much less anisotropic and the computed Fermi velocity has the highest common value $\approx 1.2 \times 10^6$ m/s along the Γ–Y line. The distinct difference in the dimensionality of the FS's between $LaSb_2$ and $LaAgSb_2$ compounds

may be responsible for inducing the SC state in the former system owing to the topological Feshbach shape resonance of the FS sheets [26]. The more 3D character of the FS sheets in $LaSb_2$ can provide even an interband scenario of the electron SC pairing, in analogy to the isostructural $LaTESb_2$ (where TE is transition element) superconductors which may result in the slightly enhanced value of T_c [16]. This is in contrast to the case of $LaAgSb_2$, exhibiting no SC state, which possesses a strongly 2D character of its FS sheets.

4. Conclusions

We have investigated electronic structures of $LaSb_2$ and $LaAgSb_2$ systems with scalar and fully relativistic DFT methods. Both the analysis of weighted bands and an exclusion of the possibility of the carriers compensation mechanism strongly suggest that the unsaturated linear MR in $LaSb_2$ is caused by the quantum limit of the Dirac fermions with high Fermi velocities. Furthermore, the small effective masses may reflect the Dirac-like dispersion of electronic bands near E_F that are only slightly modified by SOC near band crossing points. The comparison between the presented bulk Dirac states and possible topologically non-trivial surface states of the same compounds would be particularly interesting due to the bulk–boundary correspondence. However, it is planned to be a subject of further research, in analogy to the well known bulk Dirac system, ZrSiS [22], which also has unusual 2D Dirac states.

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