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Fermi surface of LaSb₂ and direct observation of a CDW transition

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Abstract

We have studied the temperature-dependent electronic structure of the light rare-earth antimonide LaSb₂ by combining angle-resolved photoemission (ARPES) measurements and density functional theory (DFT) calculations. ARPES measurements show the appearance of band replicas in the low-temperature Fermi surface with $q = 0.25 \pm 0.02 \text{ \AA}^{-1}$ and a band folding of the same periodicity with a considerable spectral weight in the folded band. From our calculations we elucidate that the folded band exhibiting the new periodicity is associated with the La-Sb layer. We propose that an in-plane distortion, probably affecting the whole unit cell, is the structural modification related to the electronic changes. All these results converge to support that a charge density wave is stabilized in LaSb₂ at low temperature.

Keywords: Layered material, Charge Density Wave, Angle-resolved photoemission, Density Functional Theory

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Charge Density Waves (CDW) have received ample attention and have been actively searched during many years due to their complex and interesting phenomenology [1]. The CDW is a collective state with a periodic symmetry-lowering charge redistribution. The periodicity is determined by the electronic structure, which is modified once the CDW is stabilized, so that the total electronic energy is decreased. The CDW is accompanied by a periodic lattice distortion [2]. It competes sometimes or even coexists with superconductivity in a process not yet well understood [3-8].

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The concept of CDW was introduced by Peierls for a one-dimensional metal [2,9]. It requires a periodic lattice distortion of $2k_F$ periodicity (Peierls distortion) and the opening of a band gap at k_F . The Peierls model involves the softening of a phonon related to the periodic lattice distortion, because the one-dimensional response function will peak at $2k_F$, as electrons will screen phonons of this wave vector very efficiently. The one-dimensional Peierls model has been extended to more complex low-dimensional systems using the concept of “nesting”, i.e. quasi-1D portions of the Fermi surface that would match after following a rigid displacement in reciprocal space. In this case only partial gapping and nesting would be expected. The experimental phenomenology is complex and the interpretation of the results has been found to be very difficult in many real systems that present a CDW, [10]. For instance, a periodic lattice distortion may appear without apparent changes in the electronic structure [11,12] or the detection of a soft phonon may be elusive [13,14]. In fact, the relevance of Fermi surface nesting to explain charge-ordering transitions has been criticized from different points of view. It has been pointed out that more complex mechanisms that go beyond the Peierls instability have to be invoked in real systems that exhibit a CDW [15], notably the momentum dependence of the electron-phonon interaction [16], i.e. a concerted action of electronic and lattice subsystems [17], and also other phenomena, like excitonic condensation [18] or electron correlations [19]. Extended first-principles calculations have shown that the CDW instabilities may have different physical origins, including the Peierls model [20] but also other mechanisms [21,22].

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Layered materials are excellent model systems to investigate these phenomena because they are prone to present CDWs [23,24]. An example of this kind of materials are light rare-earth dantimonides RSb_2 ($R=La-Nd$). This family of compounds crystallizes in the $SmSb_2$ structure, space group $Cmce$ (64), with alternating layers of Sb and RSb (see Fig. 1) and has a rich and complex physical phenomenology. While $CeSb_2$, $PrSb_2$ and $NdSb_2$ present magnetically ordered states, $LaSb_2$ is non-magnetic. It becomes superconducting at 0.4 K [25] although the onset of the transition is at 2.5 K. The emergence of coherent coupling between layers is believed to be the origin of the observed 2D to bulk transition within the superconducting regime [26].

71 The magnetoresistance of LaSb₂ exhibits a puzzling behavior. It is very large
72 (10 000%) and linear at least up to fields of 45 T [27]. Both its magnitude and its
73 linearity are at odds with the standard semi-classical theory of magnetoresistance
74 [27,28] that predicts a quadratic dependence with magnetic field, except for open
75 Fermi surface orbits, where a saturation for high fields is expected. Several
76 mechanisms may explain a linear magnetoresistance, like local fluctuations in
77 carrier density [29], high field quantization effects [30], disorder or
78 inhomogeneities [31], or magnetic breakdown effects in CDW materials [32].
79 Carrier density fluctuations and quantization effects are predicted to be
80 measurable only for very low carrier concentrations in extremely high mobility
81 samples, conditions that LaSb₂ does not meet. Moreover, the magnetization of
82 LaSb₂ displays quantum oscillations [33], and this discards most sources of
83 inhomogeneity and disorder. Finally, linear magnetoresistance has been
84 observed in some materials hosting Dirac fermions [34]. This mechanism has
85 been invoked as the reason for linear magnetoresistance in LaAgSb₂, a
86 compound related to LaSb₂. In turn, LaAgSb₂ is not superconductor, at least down
87 to 0.5 K [35].

88 A CDW may also be the origin of a linear MR, as observed for the prototypical
89 CDW layered compounds NbSe₂ and TaSe₂, which display large linear MRs [36].
90 A CDW could be present in LaSb₂, as energy band structure calculations [33]
91 predict significant nesting between the Fermi surface sheets that produce
92 experimental open orbits, which cannot be detected in de Haas-van Alphen
93 experiments [33]. Furthermore, related compounds like LaAgSb₂ [37] or
94 La_{1-x}Ce_xSb₂ [38] exhibit CDW ordering. Due to these reasons, a CDW in LaSb₂
95 has been actively searched. The kink in the resistivity found for La_{1-x}Ce_xSb₂ in a
96 broad range of Ce concentrations was considered the evidence of a CDW in this
97 solid solution [38]. The observation of a correlated kink in LaSb₂ at 355 K would
98 point to the formation of a CDW in this material as well [36]. However, STM
99 experiments did not find clear evidence of a CDW phase at 0.15 K [39]. Thus,
100 neither previous photoemission experiments at 140 K [40] nor STM experiments
101 at 0.15 K [39] have found indications of CDW formation. Finally, Fisher and
102 coworkers [41] have proposed that defects in the crystalline structure (twinning
103 or stacking faults) could be behind the CDW state and the unsaturating linear
104 magnetoresistance.

105 In this work, we report a temperature-driven phase transition that modifies
106 the Fermi surface and the band structure, leading to the appearance of band
107 replicas at low temperature associated with a reciprocal lattice vector q related to
108 k_F . Density functional theory (DFT) calculations on distorted and undistorted
109 structures allow to interpret and understand the experimental results and explain
110 them by the stabilization of a CDW.

112 **Experimental and calculation details**

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2 113 LaSb₂ single crystals are grown from high purity La and Sb by the metallic
3 114 flux method [42]. The experiments are carried out in an ultrahigh vacuum (UHV)
4 115 chamber with a base pressure below 10⁻¹⁰ mbar. Angle-resolved photoelectron
5 116 spectroscopy (ARPES) is performed using a Scienta R4000 electron analyzer
6 117 ($\pm 15^\circ$ acceptance angle) at the Cassiopeé beamline of SOLEIL synchrotron
7 118 radiation laboratory. The base energy resolution is better than 1 meV with 0.1°
8 119 angular resolution. ARPES experiments are performed after in-situ cleaving the
9 120 samples under ultrahigh-vacuum conditions. The sample cleaves between the
10 121 La/Sb bilayer, exposing a surface that it is composed by (001) Sb atomic planes.
11 122 The quality of the cleavage is good in regions much wider than the photon beam.

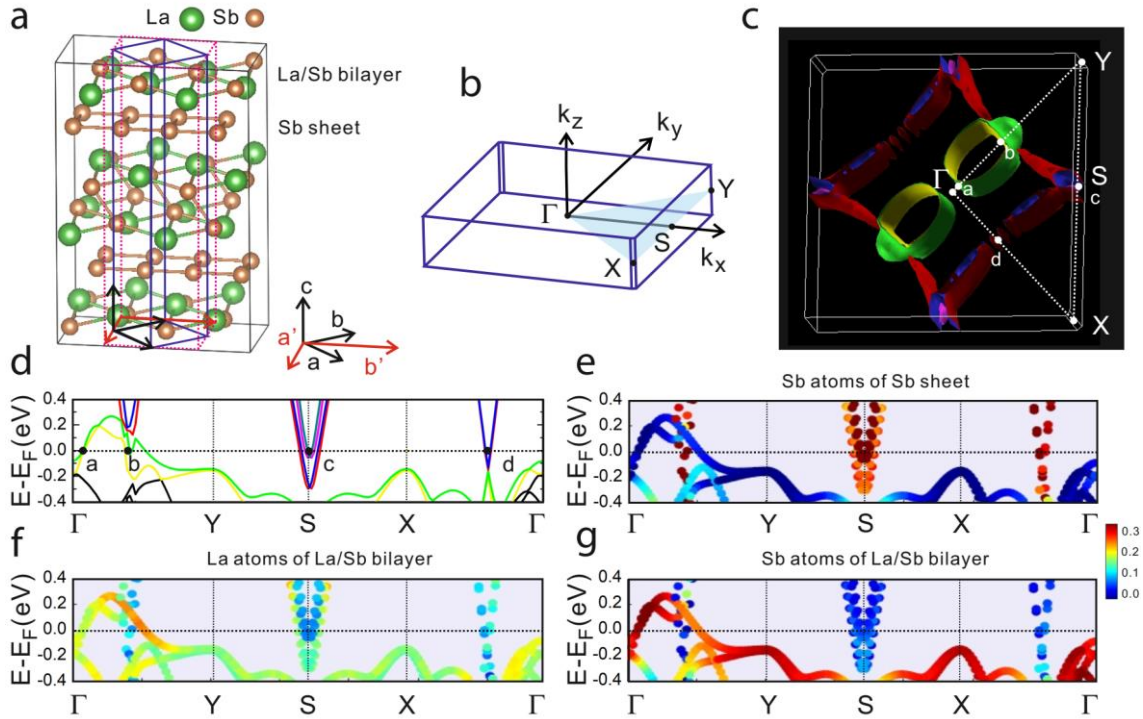
12 123 Density Functional Theory (DFT) electronic band calculations are carried out
13 124 [43] by combining the Vienna ab-initio simulation package (VASP) [44,45] and
14 125 post-processing with the VASPKIT package [46]. The Generalized Gradient
15 126 Approximation (GGA) in the Perdew, Burke, Ernzerhof (PBE) formulation [47] is
16 127 used for the exchange-correlation potential. The cutoff energy is set to 500 eV. A
17 128 Monkhorst-Pack [48,49] k-meshes with a size of 8×8×2 for the LaSb₂ primitive
18 129 cell and a size of 8×2×2 for the LaSb₂ supercell are used for the geometry
19 130 optimization. The self-consistent field convergence for the total energy and the
20 131 force variation are set to 10⁻⁶ eV and 0.001 eV/Å, respectively.

30 132 31 133 **Results and discussion**

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33
34 134 Fig. 1a shows the crystalline structure of LaSb₂ (a=b=4.483 Å, c= 18.671 Å).
35 135 The structure presents alternating Sb 2D sheets and La/Sb bilayers, so that Sb
36 136 2D sheets are sandwiched between the corrugated La/Sb bilayers along the c
37 137 axis. Fig. 1a shows the primitive cell, together with a conventional unit cell, more
38 138 convenient for visualizing the structure. The corners of the square basis of the
39 139 primitive cell are located in the middle points of the sides of the square basis of
40 140 the conventional cell on the (001) surface. Fig. 1b shows the Brillouin zone of the
41 141 LaSb₂ primitive cell and the k_x, k_y, k_z axis corresponding to the a, b, c axis in real
42 142 space in Fig. 1a, respectively.

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47 143 We have calculated the electronic structure of LaSb₂ and the results are
48 144 shown in Figs. 1c-g. Fig. 1c presents the 3D Fermi surface. The Fermi surface
49 145 consists of two main pieces: a large diamond (warped square) and two glasses-
50 146 like pockets. Unlike other compounds of the same family, like LaAgSb₂ [50], in
51 147 LaSb₂ there are no Fermi surface sheets centered on the $\bar{\Gamma}$ point [51]. The two
52 148 glasses-like pockets run along the $\bar{\Gamma} \rightarrow \bar{Y}$ direction and each of them contains two
53 149 sheets (yellow and green in Fig. 1c). The warped square contains also two sheets
54 150 (blue and red in Fig. 1c) that extend to the edges of the Brillouin zone, with
55 151 corners that are located at \bar{S} points. Four additional small pockets (purple in Fig.
56 152 1c) are located at the \bar{S} points. The Fermi surface sheets found are in agreement

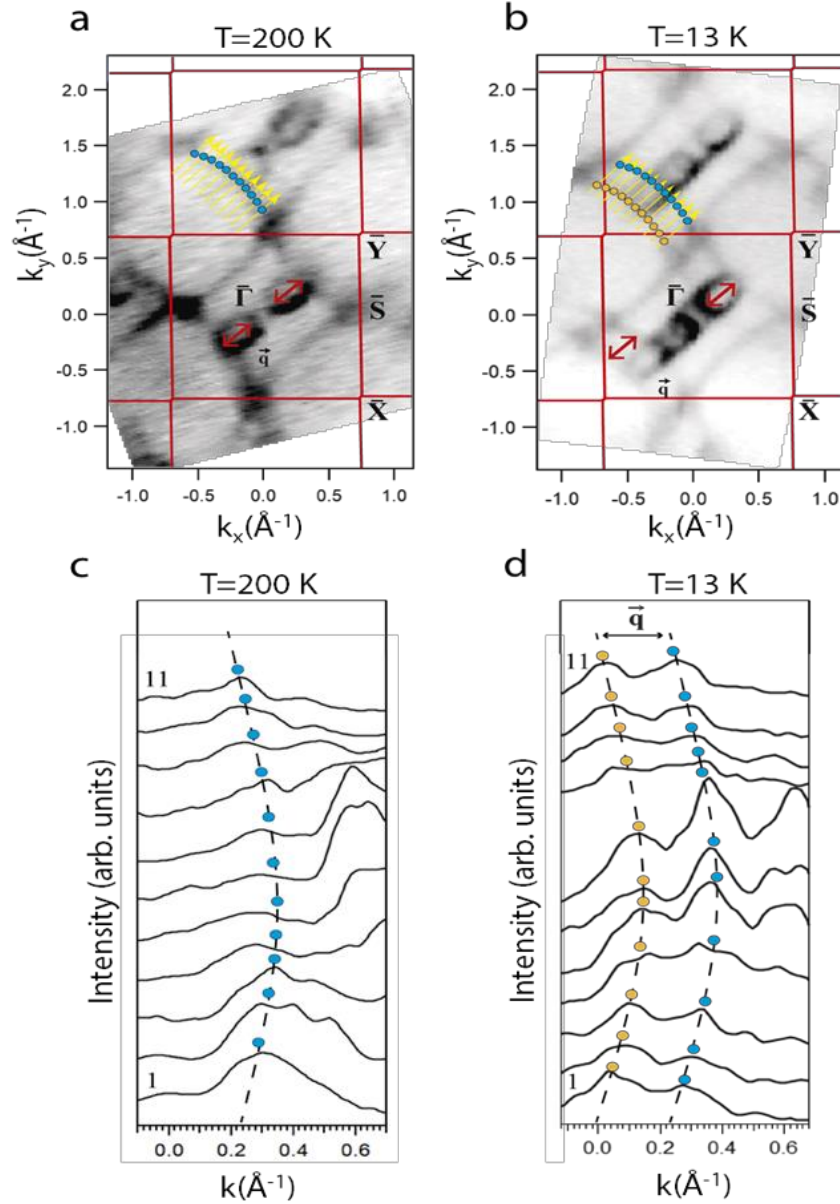
153 with previous calculations and with results from de Haas-van Alphen experiments
 154 [33,52]. Furthermore, based on the Fermi surface image, we can observe that the
 155 dispersion along k_z is small, in agreement with the quasi two-dimensional nature
 156 of the electronic structure of a layered material.



157
 158 Fig. 1 (a). Layered LaSb₂ crystalline structure, formed by Sb sheets and La/Sb bilayers.
 159 The primitive and conventional cells are marked with blue and red lines, respectively. b.
 160 Brillouin zone corresponding to the LaSb₂ primitive cell. (c-g) Theoretical electronic band
 161 structure of LaSb₂ c. Calculated Fermi surface of LaSb₂. The Fermi surface sheets are
 162 highlighted using different colors: yellow and green for the inner glasses-like pockets,
 163 blue and red for the outer warped square and purple for the small pockets near \bar{S} points.
 164 d-h. Electronic band structures of LaSb₂ along Γ -Y-S-X- Γ K paths. The band structure is
 165 projected on the Sb atoms of the Sb-sheets (e), La atoms of the La/Sb bilayers (f), and
 166 Sb atoms of the La/Sb bilayers (g). The color scale indicates the contribution for each
 167 band at the different atoms (red is higher). The crossing points of the bands with the
 168 Fermi energy (a,b,c,d) are roughly marked in panels c and d.

169
 170 The electronic band structure along Γ -Y-S-X- Γ K paths is shown in Fig.1d-g.
 171 The energy range of the calculated band structure is restricted to values around
 172 the Fermi energy in order to compare the calculations with experimental bands
 173 more clearly (see below). Fig.1d shows the bands crossing the Fermi level
 174 (crossing points marked by a,b,c,d). In order to better relate the band structure
 175 and the Fermi surface, these crossing points are also indicated in the Fermi
 176 surface shown in Fig.1c. Further information on the Fermi surface sheets is
 177 extracted from the atomic origin of the bands. Figs. 1e-g show the atom-projected
 178 band structures. Fig. 1e corresponds to projection on the Sb atoms of the Sb-

179 sheets, Fig. 1f on the La atoms of the La/Sb bilayers, and Fig. 1g on the Sb atoms
180 of the La/Sb bilayers. Note that the warped square sheets of the Fermi surface
181 have a strong Sb content from atoms in the Sb sheets (Fig. 1e). These bands
182 present quasi-conical dispersion near the \bar{S} points (Fig. 1e). The two glasses-like
183 pockets in the Fermi surface arise mainly from the La and Sb atoms in the La/Sb
184 bilayers, as shown in Figs. 1f and 1g. A close inspection of the Fermi surface
185 shown in Fig. 1c reveals sheets that are parallel to each other, susceptible to nest
186 and therefore favorable to induce a CDW instability, as previously suggested [33].
187 Nesting implies that significant parts of the Fermi surface are connected by a
188 wave vector q [9]. In this case, replicas of the Fermi surface sheets appear when
189 the CDW sets in [53,54]. In the search for changes in the electronic structure of
190 LaSb₂ related to a CDW, which have been elusive up to now, we have performed
191 ARPES measurements covering an extended area of reciprocal space. The
192 intensity at the Fermi level is obtained by integrating the spectral weight in a
193 window of ± 20 meV around the Fermi energy. Fig. 2a, b show the Fermi surface
194 of LaSb₂ at T=200 K and T=13 K respectively (a larger region of the reciprocal
195 space is shown in the Supplementary Material) for $h\nu = 118$ eV. At 200 K the
196 shape of the experimental Fermi surface reproduces the warped square and the
197 glasses-like pockets. When the temperature is lowered to T=13 K, the shape of
198 the Fermi surface changes dramatically. As seen Fig. 2b, the Fermi surface is
199 replicated along the $\bar{\Gamma Y}$ direction, the same direction along which the glasses-like
200 pockets are oriented. In order to analyze these changes in detail, we have made
201 several cuts in the Fermi surface parallel to this direction. The cuts have been
202 made both perpendicular to the outer edge of the warped square (yellow arrows
203 in Fig. 2a, b) and along the glasses-like portions (Fig. S1). These cuts (labelled
204 with numbers) are represented in Fig. 2c, d. The maxima of the intensity are
205 highlighted with colored dots and the location of the maxima in reciprocal space
206 is shown in Fig. 2a, b. While at T=200 K only one maximum appears, at T=13 K
207 a replica is observed. The separation between the two maxima is constant and
208 corresponds to a value of $q = 0.25 \pm 0.02 \text{ \AA}^{-1}$ (equivalent to $(0.18 \pm 0.01) 2\pi/a$).
209 The Supplementary material shows additional analysis of Fermi surface regions
210 interconnected by this q vector.

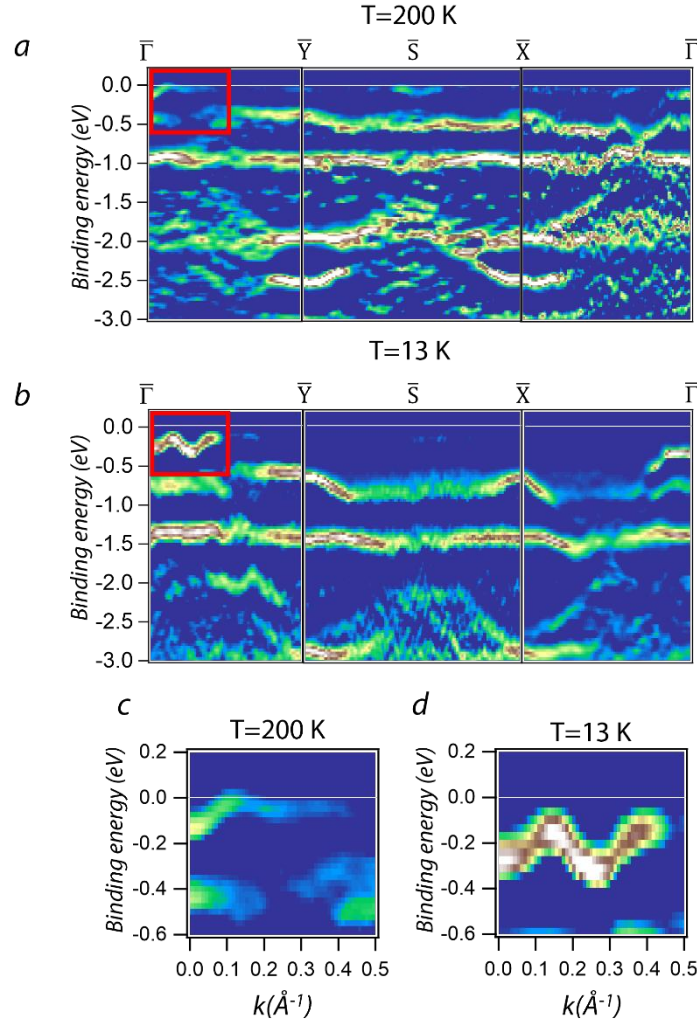


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212 Fig. 2. Experimental k -space mapping of the Fermi surface obtained from the plot of the
 213 ARPES intensity as a function of the k_x and k_y vectors near the Fermi level ($h\nu= 118$ eV).
 214 The measurements are done at (a) $T=200$ K and (b) $T=13$ K. The panels cover the first
 215 and second Brillouin zones. Yellow arrows indicate the places where cuts of the Fermi
 216 surface are done. The cuts corresponding to the second Brillouin zone, labeled by
 217 numbers, are represented in panels (c) and (d) respectively, where $k=0$ corresponds to
 218 the arrow start. The blue and yellow dots in these panels indicate the intensity maxima.
 219 While at high temperature there is only one maximum, i.e., one band crossing the FL
 220 (blue dots), at low temperature there are two bands (yellow dots). The maxima are
 221 separated by a distance $q= 0.25 \pm 0.02 \text{ \AA}^{-1}$. This vector (length and direction) is depicted
 222 in panel (b).

223

224 We present the electronic structure measured by ARPES along the high
225 symmetry directions of the Brillouin Zone ($\bar{\Gamma}\bar{Y}\bar{S}\bar{X}\bar{\Gamma}$) in Fig. 3, measured at 200 K
226 and at 13 K. After cleaved, the exposed surface is composed by (001) Sb atomic
227 planes (Fig. 1a). Fig. 3 represents the curvature plot [55] of the ARPES intensity
228 obtained with a photon energy of $h\nu=118$ eV on a wide range of binding energy.
229 The curvature analysis is a powerful method to enhance dispersive features in a
230 spectroscopic image and is typically used to highlight the region near the Fermi
231 level, whose intensity may be weak in comparison with the rest of the band
232 structure. There are clear modifications as a function of the temperature in the
233 high binding energy bands. At high temperature, there are two low-dispersing
234 bands around -0.5 eV and -1.0 eV, whose energy difference increases at low
235 temperature. The observed changes in binding energy and dispersion might be
236 attributed to band Umklapps and/or to increased correlation effects in the low
237 temperature phase, but further work is needed to discriminate the origin.
238 However, the most important difference between the data at 200 K and at 13 K
239 is seen in the Fermi level region, where electronic instabilities are relevant. In the
240 neighborhood of $\bar{\Gamma}$ along the $\bar{\Gamma}\bar{Y}$ direction, the bands present a strong folding at
241 13 K. This modification of the band structure corresponds to the glasses-like
242 pockets associated with the La-Sb bands (red rectangle in Fig. 3a, b). This region
243 is enlarged in Fig. 3c, d, which shows how these bands are folded at low
244 temperature. The spectral weight in the folded band is very strong, indicating that
245 a strong potential is inducing the folding. Simultaneously to the band folding, it
246 seems to be a weight reduction in the lowest binding energy states at low
247 temperature. A spectral weight reduction in the whole Brillouin zone is compatible
248 with Mott transitions, that can be triggered by the bandwidth reduction W due to
249 CDW band folding, since the W/U ratio (U being the electronic correlation) is more
250 favorable for Mott physics [12]. Figure 3 does not exhibit gap opening. The
251 intensity decreases at the Fermi energy associated to the band gap of the CDW
252 can only be observed when the CDW gap is larger than the width of the bands.
253 Not observing the band gapping at the Fermi surface only means that the band
254 gap is possibly smaller than the width of the electronic bands (here around 100
255 meV). Since the transition temperature is somewhere between 200 K and 13 K,
256 the maximum band gap could be associated to 200 K, i.e., 18 meV. Therefore, in
257 this case the intrinsic width of the electronic bands does not allow us to observe
258 the band gap at the Fermi surface.



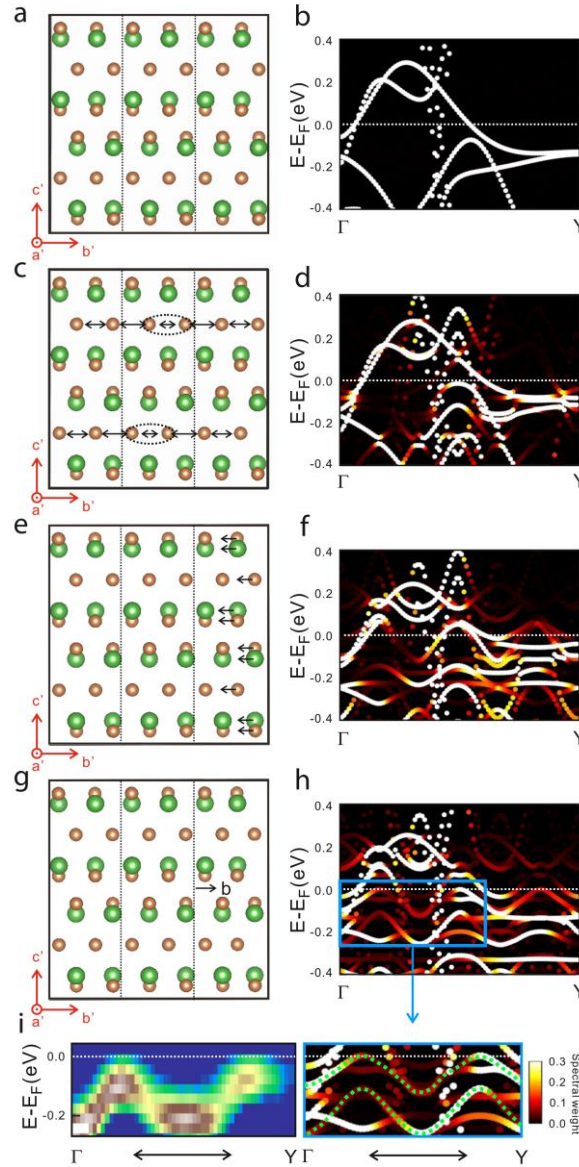
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260 Fig. 3 Experimental band structure along high symmetry directions ($\bar{\Gamma}\bar{Y}\bar{S}\bar{X}\bar{\Gamma}$) at (a) 200 K
 261 and (b) 13 K. Panels (c) and (d) are zooms of the areas indicated with a red rectangle in
 262 panels (a) and (b) respectively.

263

264 The experimental results at low temperature show both replicas at the Fermi
 265 surface and a clear folding of a La-Sb band. Replicas are connected to the
 266 original spectral features by a reciprocal space vector of the size of the La-Sb
 267 pocket. This corresponds to one-third of the $\bar{\Gamma}\bar{Y}$ distance, so that the periodicity of
 268 the atomic structure in real space should be 3 times larger along the \mathbf{b}' direction
 269 (i.e., $3\mathbf{b}'$, see Fig. 1a). These electronic changes are compatible with the
 270 stabilization of a CDW at low temperature (13 K). The data at 200 K reflect the
 271 electronic structure of the high temperature phase (normal phase). In order to
 272 understand the nature of the structural distortion associated with this wave vector,
 273 we have constructed a supercell of size $3\mathbf{b}'$, i.e. it contains 3 conventional cells
 274 along the \mathbf{b}' direction, since this size will allow us to describe the observed q
 275 vector. Fig. 4a shows a supercell with no structural modification (called hereafter
 276 *pristine* supercell) and corresponding to the high-temperature phase. In order to

277 obtain an effective primitive cell band structure, unfolded band structures [56] are
 278 calculated for the supercells studied. Fig.4b shows the unfolded band structure
 279 of the pristine supercell, where we observe that the bands are in reasonable
 280 agreement with the bands of the primitive cell (Fig.1).



281

282 Fig. 4. Structural modifications and the corresponding band structures. (a)
 283 Supercell of LaSb₂ containing 3 conventional cells along **b'** direction. (b)
 284 unfolded band structure of the supercell in (a). (c), (e), (g). LaSb₂ supercells with modified
 285 atomic positions to simulate the periodicity of the CDW: (c) Displacement of the
 286 Sb atoms in Sb sheets along **b'** direction to introduce a longitudinal-wave-like
 287 modification in the Sb sheet. (e) Displacement of half of the atoms in the
 288 conventional unit cell (those in the right conventional cell). (g) Displacement of
 289 the whole middle conventional cell by 0.02 Å along the **b'** direction. (d), (f), (h).
 290 Unfolded band structures correspond to the modified supercells in (c), (e), and
 291 (g), respectively. (i) Area near E_F from the experimental band structure (Fig. 3b)

292 and theoretical band structure of an atomic distortion affecting the whole unit cell
293 (h).

294

295 Considering that this supercell describes the electronic structure at 200 K
296 (high-temperature phase), we focus on describing the low-temperature phase.
297 The experimental results show a clear folding of the glass-like features,
298 corresponding to one-third of the $\bar{\Gamma}-\bar{Y}$ distance, i.e. $3\mathbf{b}'$ in real space or
299 equivalently 3 times along the $\mathbf{a}+\mathbf{b}$ direction. This information is hard to obtain
300 straightforwardly from DFT. As DFT calculations are static simulations,
301 temperature-dependent changes are difficult to simulate [57]. Therefore, in order
302 to calculate the low-temperature crystal structure, we have forced different
303 structural modifications in the pristine supercell. The displacements correspond
304 to the experimental \mathbf{q} vector of the CDW. This procedure induces a CDW in the
305 system corresponding to the experimental \mathbf{q} vector. We have analyzed several
306 structural modifications with a $3\mathbf{b}'$ periodicity. In particular, we have introduced
307 displacements (1) on the Sb sheets, (2) on part or (3) on all of the atoms of one
308 conventional cell. Fig.4c shows a longitudinal-wave-like modification in the Sb
309 sheets (method 1), where Sb atoms in Sb sheets are displaced along \mathbf{b}' direction.
310 The two middle Sb atoms in the Sb sheets are displaced towards each other
311 along \mathbf{b}' direction, reducing their distance by 0.03 Å. Then we fix these atoms,
312 and the positions of the rest of the atoms are relaxed. Fig. 4e shows the
313 displacement of half of the atoms in the conventional unit cell. Atoms on the right
314 side of the unit cell are shifted by 0.02 Å along \mathbf{b}' direction. Then, the right-part
315 of the conventional cell is fixed and again all the other atoms are relaxed (method
316 2). Fig. 4g shows the displacement of the whole middle conventional cell along \mathbf{b}'
317 direction by 0.02 Å (method 3). This represents a wave like distortion along \mathbf{b}' .
318 Furthermore, we have also tried many other structural modifications, such as
319 displacement of the Sb atoms in Sb sheets by 0.03 Å along \mathbf{a}' direction,
320 displacement of the Sb atoms inside the right-side conventional cell 0.02 Å and
321 0.04 Å along \mathbf{b}' direction, and displacement of the whole middle conventional cell
322 by 0.02 Å along the \mathbf{a}' direction and \mathbf{c}' direction, which are shown in the
323 Supplementary Material. The unfolded band structures of these different modified
324 structures are shown in Fig.4 and Fig.S4.

325 We want to stress that not every structural modification of the periodicity
326 reproduces the experimental band structure. We also observe that the distorted
327 structures introduce more bands, compared to the unfolded band structure of the
328 pristine supercell. Band replicas are observed for some structural deformations,
329 such as Fig.4d,h and Fig.S4d,f,j. In particular, the displacement of the whole
330 conventional middle cell along \mathbf{b}' direction (method 3)) shows more clearly band
331 replicas and also the band shapes are almost preserved (Fig.4h). A detailed
332 comparison of the experimental and calculated band structures with this
333 structural distortion are shown together for comparison in Fig.4i. There is a fair

334 agreement with the experiment. Note the presence of band replicas just below
335 the Fermi level with a dispersion similar to the experimental bands. This rough
336 model captures the essential physics of the system, so the temperature-driven
337 phase transition of LaSb₂ is traced back to a periodic distortion of the structure
338 along the **b'** direction, involving most of the atoms of the conventional unit cell.
339 The atomic displacements introduced are in the range of 0.02-0.03 Å. For much
340 smaller displacements, band replicas are hard to observe, and for much higher
341 displacements, the band structure deviates seriously from the original one (see
342 for instance Fig.S4f). Since the experimental low temperature bands and Fermi
343 surface keep the original shape, except for the replicas, the structural
344 modifications cannot deviate much from the above-mentioned values. We
345 conclude that the displacements that originate de CDW are roughly in the range
346 of 0.02-0.03 Å.

347 It is interesting to compare the periodicity of the CDW observed at low
348 temperature with the periodicities identified previously for the related compound
349 LaAgSb₂. We would like to point out that this comparison should be made with
350 care, because the two compounds have a closely related structure and
351 composition, although the shape of their Fermi surfaces is different, in particular
352 the number and shape of the Fermi surface sheets are not the same. The CDW
353 of LaAgSb₂ presents a periodic lattice distortion with a periodicity corresponding
354 to $0.026 \frac{2\pi}{a}$ [37], much smaller than the value of $0.18 \frac{2\pi}{a}$ found in this work for
355 LaSb₂. In turn, the small value found for LaAgSb₂ hinders the observation of
356 modifications in the electronic structure. In fact, the effects related to the
357 stabilization of the CDW are limited to a reduction of intensity in the region close
358 to the Fermi energy [58]. An analysis of the region close to the Fermi energy in
359 LaSb₂ shows the strong band folding observed in Figs. 3c,d. It is interesting to
360 analyze the behavior in the rest of the reciprocal space as well. The observation
361 of folding in Fig. 3c,d does not involve gapping, as shown by the theoretical
362 folding depicted in Fig. 4i and evidenced by the observation of replicas in the
363 Fermi Surface in Fig. 2b. We note that the less intense bands near the Fermi level
364 area are hardly seen in the curvature plot of Fig. 3b. If we want to compare the
365 intensity of the bands near the Fermi energy for the high temperature phase (200
366 K) and the CDW phase (13 K), we need to resort to the original data, as the
367 curvature plot may induce a relative change of intensity of the bands. This
368 analysis is made in the Supplementary Information, where Fig. S2 and S3 show
369 an intensity reduction near the Fermi energy, without any apparent gapping, as
370 observed for LaAgSb₂, and in agreement with de Haas-van Alphen
371 measurements [33], which did not find Fermi surface sections affected by a
372 possible CDW at low temperature.

373 Finally, it is worth to compare these findings with the results for La_{1-x}Ce_xSb₂
374 [38] as well. The substitution of La by Ce generates a pronounced kink in the
375 resistivity vs. temperature, also observed in the specific heat, suggestive of the

376 formation of a CDW. An analogous kink in the resistivity for LaSb₂ would
377 correspond to a CDW in this material at 355 K, i.e. well above the temperature
378 range found in this work. However, sequential CDW distortions are not rare, as
379 they have been observed in several compounds, like the very closely related
380 compound LaAgSb₂ (207 K and 16 K), and in other compounds such as 2H-
381 TaSe₂ (122 K and 90 K), 1T-TaS₂ (1120 K, 350 K, 180 K), NbSe₃ (145 K, 59 K)
382 [12,32,37,59-61]. As no specific periodic lattice distortion was reported for La_{1-x}
383 Ce_xSb₂, it is not possible to establish any relationship between the CDW at 355
384 K and the CDW found here below 200 K and experimentally observed at 13 K,
385 although sequential CDW distortions have been observed in several compounds
386 [12,32,37,59-61].

387 In summary, ARPES measurements reveal the presence of changes in the
388 electronic structure of LaSb₂ surface between 200 K and 13 K, including band
389 folding and replicas in the Fermi surface. All these modifications are signatures
390 of the stabilization of a CDW appearing below 200 K and observed at 13 K, above
391 the superconducting critical temperature of the system (0.4 K). The changes in
392 the electronic structure at low temperature are related to the same q vector ($q =$
393 $0.25 \pm 0.02 \text{ \AA}^{-1}$), indicating that they correspond to an intrinsic property of the CDW
394 phase. The q vector found connects regions inside the glasses-like pockets of the
395 Fermi surface. The results of theoretical calculations permit to interpret these
396 findings. The calculations show that the glasses-like pockets involved in the CDW
397 transition exhibit La-Sb character and that the transition corresponds to a small
398 structural distortion affecting all the atoms in the unit cell. We also propose that
399 the CDW transition appearing at 13 K could trigger a Mott transition at lower
400 temperatures due to the bandwidth reduction, as in other systems, a scenario
401 worth exploring.

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