Substrate-mediated umklapp scattering at the incommensurate interface of a monatomic alloy layer

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I. INTRODUCTION

Two-dimensional (2D) materials have been a research focus in recent years. 2D-xenes such as silicene [1,2], germanene [3,4], stanene [5,6], borophene [7,8], bismuthene [9], and transition metal dichalcogenides monolayers (TMD), such as MoS$_2$ [10], WS$_2$ [11], MoSe$_2$ [12], WSe$_2$ [13,14], MoTe$_2$ [15], are considered as 2D topological insulators and 2D semiconductors with major potential for industrial applications. For TMD, the bonding is mainly van der Waals type so the epitaxial lattice grows with the lattice constants of their bulk structure in spite of large lattice mismatch at the interface. The condition for epitaxial growth of xenes on substrates is stricter than the commensurate interface suggested to be necessary [16], and the growth configurations are more varied. For example, germanene was found to have dual phases grown on Ag(111), that is, a striped phase (SP) and a quasifreestanding phase (QP) [3]. The former is uniaxially commensurate with the Ag(111)-$\sqrt{3} \times \sqrt{3}R30^\circ$ lattice (hereafter denoted as Ag$_{\sqrt{3} \times \sqrt{3}R30}$) and exhibits a well-ordered but tensile-strained honeycomb lattice; the latter is incommensurate with Ag(111) and reveals a twisted or imperfect honeycomb lattice. However, the intrinsic $\sigma$ band of germanene was only observed in the latter [3]. Therefore, commensurability is not a necessary condition for growing a monoatomic layer on a substrate, and incommensurability can even preserve better the intrinsic electronic structures, such as $\sigma$ bands, of a monolayer [3] due to a less effective interaction with the substrate. However, the $\pi$ bands appear more vulnerable to the interaction with the substrate regardless of the commensurate or incommensurate interface [3,17,18]. The stability of a film was mainly investigated in terms of surface energies with a model of a freestanding slab, while the interfacial effect from the substrate was considered as a secondary factor [19]. However, when the film thickness reduces to one monolayer, the interfacial effect should become more relevant. Tang et al. [20] found that Pb films grown on Ge(111) are dominated by the configuration with 13% lattice mismatch at a thickness less than 2 ML. This was attributed to the stronger electron hybridization at the interface between the Pb film and Ge(111) substrate.

In this paper, we focus on surface alloys of Ag$_2$Ge [21,22] and Ag$_2$Pb [21,23] first, which are actually single alloy layers forming on the bulk Ag(111) surface. From the distinct features of electronic structures between the two surface alloys, mainly the splitting and nonsplitting surface state bands centered at $\bar{M}_{\text{Ag}(111)}$, we correlate this presence or absence of the band splitting to the corresponding incommensurate and commensurate interfaces of these two systems. Upon further deposition of Pb on Ag$_2$Pb/Ag(111), a dealloying process occurs; the commensurate Ag$_2$Pb/Ag(111) evolves into an incommensurate 1-ML dense Pb(111) layer on Ag(111) with its Pb band splitting at $\bar{M}_{\text{Ag}(111)}$, as well. The substrate-mediated umklapp scattering of the monolayer electrons at the interface is proposed to explain the observed band split and help the layer stability despite incommensurability.
II. CALCULATIONAL METHODS AND EXPERIMENTAL PROCEDURES

The ab initio calculations are performed based on density functional theory (DFT) with the Vienna Ab initio Simulation Package (VASP) [24–26]. The Perdew-Burke-Ernzerhof [27] generalized-gradient-approximation functional and projector augmented wave [28,29] method are adopted in the self-consistent calculations. Two calculation approaches were employed in this paper: (1) The model of surface alloys on Ag slabs: The \( \text{Ag}_2\text{Ge} \) or \( \text{Ag}_2\text{Pb} \) alloy was formed on the double side surfaces with a repeating slab of 20 Ag layers and a vacuum region of \( \sim 26 \text{ Å} \). The theoretical optimized lattice constant of 4.146 Å for the Ag slabs was used, and Ge, Pb, and Ag atoms were allowed to fully relax except that the four Ag layers in the middle were fixed at their bulk positions. The plane-wave kinetic cutoff energy was 300 eV and the force on each atom was less than 0.005 eV Å\(^{-1}\). (2) The model of freestanding layers: The energy cutoff of 400 eV (400 eV) and \( k \) mesh of \( 12 \times 12 \times 1 \) (24 \( \times 24 \times 18 \)) were used in free-standing \( \text{Ag}_2\text{Ge} \) and Pb-1ML (Ag bulk). The DFT + Hubbard \( U \), local-density approximation + \( U \), method [30] with \( U = 4.0 \) eV and \( J = 0.4 \) eV [3] are applied on the Ag-\( d \) orbital to correct the binding energy of the Ag-\( d \) bands in \( \text{Ag}_2\text{Ge} \) and Ag bulk. To simulate the influence of umklapp scattering to the electronic bands, the band structures of \( \text{Ag}_2\text{Ge} \) and Pb 1-ML were unfolded back to the high-symmetry lines in the Ag(111) first Brillouin zone (See Supplemental Material Fig. S1 [31]). We also simulate the low energy electron diffraction (LEED) result of the Ag(111) surface with 2D hexagonal lattice parameter 2.88 Å (equal to 4.073 Å for the face-centered-cubic lattice). For the \( \text{Ag}_2\text{Ge} \) alloy, we use the lattice parameter of 4.7489 Å with a rotation angle of 30° (R30°). In comparison with Ag\( \sqrt{3}-\text{R30} \), the lattice mismatch is \(-5\%\). On the other hand, for the 1-ML Pb, the lattice parameter 3.4 Å is used.

The experiment was performed in an ultrahigh vacuum system, where angle-resolved photoelectron spectroscopy (ARPS) and LEED were used. The single-crystal Ag(111) surface was cleaned by repeated cycles of sputtering with Ar\(^+\) ions at 1.5 keV followed by annealing at 500° C until its cleanliness was verified by a sharp LEED diffraction pattern as well as the presence of a sharp L-gap Shockley surface state in the ARPS spectra. Ultrathin films of Pb and Ge were grown on Ag(111) substrates at room temperature (RT) and 100° C, respectively using an K-cell evaporator. ARPS data was collected from beamline 21B1-U9 at the National Synchrotron Radiation Research Center in Taiwan with a Scienta R4000 energy analyser using \( p \)-polarized light at 27 eV, and with Scienta R3000 energy analyzer using mainly unpolarized He-I \( \alpha \) light source at 21.2 eV. The energy and angular resolutions for the former (the latter) were 10 meV and 0.3° (30 meV and 0.4°).

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the measured energy band dispersions of \( \text{Ag}_2\text{Pb} \) and \( \text{Ag}_2\text{Ge} \) surface alloys formed on the Ag(111) surface in the symmetry direction \( \bar{\Gamma} - \bar{K} - \bar{M} \) (\( \bar{\Gamma}_{\text{Ag}(111)} - \bar{\text{M}}_{\text{Ag}(111)} \)) in terms of Ag\( \sqrt{3}-\text{R30} \) and Ag(111) surface Brillouin zones (SBZs), respectively. At the surface zone center \( \bar{\Gamma} \), as reported by previous studies [21–23], unoccupied surface states (\( S_1, S_2, S_1', S_2' \)) were identified to disperse downward crossing Fermi levels for both surface alloys; those of \( \text{Ag}_2\text{Pb} \) surface alloys were especially shown to exhibit Rashba splitting with Rashba constant 1.42 Å eV [23]. At the surface zone boundary (SZB) \( \bar{M} \), the difference between the \( \text{Ag}_2\text{Pb} \) and \( \text{Ag}_2\text{Ge} \) surface alloys is more evident that the surface state band, \( S_3' \), at about \(-1.24 \) eV of the latter is splitting rather than the single one, \( S_3 \), at about \(-0.95 \) eV in the former within the bulk projected Ag band gap. The split of the \( \text{Ag}_2\text{Ge} \) surface state band at \( \bar{M} \) was speculated to be caused by Rashba effects or structure disorder [21,22]. The nearly flat bands ranging about \(-3 \) eV in both Figs. 1(a) and 1(b) and the weak replica at about \(-2 \) eV are from Ag \( d \) electrons emitted by He-I \( \beta \) and \( \gamma \), \( h\nu = 23.09 \) and 23.74 eV. Figure 1(c) shows the DFT calculation results for both surface
alloys based on the model of the alloy layers on Ag slabs of 22 ML. The lattices of both alloy layers are considered to be commensurate with Ag√3-R30 in the calculation. Via a close comparison, the calculated surface state bands match the measured ones at Γ and M reasonably well for PbAg₂ but disagree with the measured ones for Ag₂Ge in that the calculated S₁ surface state at Γ is yet below the Fermi level, and moreover, the calculation cannot reproduce the S₁ band split at M. It is obvious that the calculation model employed fits Ag₂Pb rather than Ag₂Ge. What is the essential difference between them and what factor was not considered in the calculation?

Figures 2(a) and 2(c) show the LEED patterns of Ag₂Pb and Ag₂Ge on Ag(111), taken at the same electron energy 20 eV. The six (1/3 1/3) spots in both patterns correspond to the Ag√3-R30 lattice. Although the spot positions of the two surface alloys are identical, the spot shapes appear different; the former [Fig. 2(b)] look like filled circles, while the latter [Fig. 2(d)] exhibit satellite-like configuration around (1/3 1/3) spots like Moirés. This implies that there is lattice mismatch between the Ag₂Ge layer and Ag(111), which was overlooked in the calculation for Fig. 1(c). A typical Ag₂M surface alloy normally assumes the underlying Ag√3-R30 lattice, especially when the foreign atom M is of a similar size to the Ag atom. The radius of Pb, Ge, and, Ag atoms are 1.8, 1.25, and 1.6 Å, respectively [32], so upon the Ag₂Pb alloy formation, a Pb atom, due to its larger size, can only be partially immersed into the top Ag layer until the alloy lattice is commensurate with Ag√3-R30. This is consistent with a previous LEED IV study showing that the top layer is corrugated such that the Pb atoms reside about 0.4 Å above the Ag atoms [33]. However, for a Ge atom, due to its much smaller size, the lattice of Ag₂Ge further contracts to a smaller value than that of Ag√3-R30. Therefore in Fig. 1(b), the right branch, with higher intensity, of the splitting bands centered at M_{Ag(111)} can be the original surface-state band of Ag₂Ge and the left branch, with lower intensity, is generated simply due to umklapp scattering via a Ag(111) reciprocal lattice. This is consistent with the formation of Moiré LEED spots as linear combinations of the reciprocal vectors of the film and the substrate [3] namely,

$$\mathbf{k} = n_1 \mathbf{G}_{Ag2Ge} + n_2 \mathbf{G}_{Ag(111)} (n_1 \text{ and } n_2 \text{ are integers})$$  \hspace{1cm} (1)

Figure 2(e) illustrates the model for explaining the energy band split at M_{Ag(111)} using substrate-mediated umklapp scattering, where M_{Ag2Ge} is the SZB of the Ag₂Ge surface alloy with the contracted lattice from the Ag√3-R30 unit cell. The solid bands centered at the momentum of $-M_{Ag2Ge}$ and $M_{Ag2Ge}$ are identical due to the inversion symmetry of the
FIG. 3. Comparison between measured and calculated electronic structures for Ag$_2$Ge surface alloy. (a) Schematics (side and top view) to illustrate the model for DFT calculation of a freestanding layer of Ag$_2$Ge. (b) SBZs and symmetry points of the Ag$_2$Ge layer (denoted in blue) and the Ag(111) surface (denoted in red). $\vec{a}_{\text{Ag}(111)} = 1.25$ Å$^{-1}$, $\vec{a}_{\text{Ag}_2\text{Ge}}$, $\vec{a}_{\text{Ag}_2\text{Ge}} = 1.31$ Å$^{-1}$, $\vec{a}_{\text{Ag}_2\text{Ge}} = 0.87$ Å$^{-1}$. (c) The calculated energy bands of freestanding Ag$_2$Ge derived from the model in (a). (d) Superimposition of DFT calculated band structures in (c) onto experimentally measured energy band dispersions in Fig. 1(b).

band structures. The dashed band translated from the one at $-\vec{M}_{\text{Ag}_2\text{Ge}}$ by umklapp scattering must be symmetrized with the band at $\vec{M}_{\text{Ag}_2\text{Ge}}$ with respect to $\vec{M}_{\text{Ag}(111)}$ simply because $\vec{G}_{\text{Ag}(111)} = 2\vec{M}_{\text{Ag}(111)}$. The value of lattice mismatch (LM) can be extracted by the magnitude of the splitting, $|\vec{M}_{\text{Ag}(111)}\vec{M}_{\text{Ag}_2\text{Ge}}|$, namely,

$$\text{LM} = \frac{a_{\text{Ag}_2\text{Ge}} - a_{\text{Ag}(111)} \sqrt{3} \times \sqrt{3} R 30^\circ}{a_{\text{Ag}(111)} \sqrt{3} \times \sqrt{3} R 30^\circ} = \frac{\vec{M}_{\text{Ag}(111)}\vec{M}_{\text{Ag}_2\text{Ge}}}{\vec{M}_{\text{Ag}(111)}\vec{M}_{\text{Ag}_2\text{Ge}}}$$

and the deduced value is $-5\%$ from the measured band split, 0.064 Å$^{-1}$, in Fig. 1(b). The corresponding Wood notations for the Ag$_2$Ge layer and Moiré pattern are $\frac{2}{3} \sqrt{3} \times \frac{19}{3} \sqrt{3} R 30^\circ$ and $19\sqrt{3} \times 19\sqrt{3} R 30^\circ$, respectively. With the corresponding lattice constants to the extracted value, the Moiré spots are reproduced using Eq. (1), as shown in Fig. 2(f). The yellow square encloses the Moiré spots around (1/3 1/3). The measured counterparts [Fig. 2(d)] are not clearly resolved. As previously reported [3], above 1/3 ML, the dealloying process starts engaging to form a complete SP at about 0.74 ML. It is possible that some precursors of SP already coexist with the Ag$_2$Ge alloy at 1/3 ML and effect the long-range order.

To further confirm this model, we employed a DFT calculation only considering a freestanding layer of Ag$_2$Ge as shown in Fig. 3(a). The fully relaxed lattice constant is 4.68 Å, 93.8% of that of an Ag$_{\sqrt{3}/2}/221a_{3-R30}$ unit cell. The corresponding lattice mismatch, $-6.2\%$, is close to that extracted from the measured band splitting. The smaller LM magnitude in the real case, Ag$_2$Ge on Ag(111), is understandable in light of the fact that the underlying Ag substrate is resistant to the contraction.
of Ag$_2$Ge from the Ag$\sqrt{3}$/3-R30 lattice. We, therefore, intentionally employed the lattice constant corresponding to $-5\%$ lattice mismatch for the band calculation of the freestanding Ag$_2$Ge layer, and then unfolded the calculated energy bands with the Ag(111) reciprocal vector [See Supplemental Material Fig. S1 [31]]. The resulting bands are superimposed onto the Ag bulk projected band continuum as shown in Fig. 3(c). The bright yellow bands, with higher intensities, are the original Ag$_2$Ge bands and the dark-blue bands, with lower intensities, are the unfolded bands. The SBZs and symmetry points of the Ag$_2$Ge layer (blue) and the Ag(111) surface (red) are indicated in Fig. 3(b). The calculated, including the original (solid curves) and the unfolded (dashed curves), and the measured bands [Fig. 1(b)] are compared in Fig. 3(d); the match is agreeable for either the $S_1'$ and $S_2'$ bands centered at $\Gamma$ or the splitting $S_1$ bands centered at $M_{\text{Ag(111)}}$. Note that the unfolded bands from the calculation match well the left branch of the measured splitting bands at $M_{\text{Ag(111)}}$ but hardly find counterparts about $\Gamma$ in the data. This is likely due to the very low intensity of the umklapp bands around $\Gamma$. The occupied bands dispersing upward from $\sim -1$ eV at $\bar{\Gamma}$ indicated by an arrow in the calculated result of Fig. 3(c) are also missing from the measured ones in Fig. 3(d). The reason will be addressed later. It is worth noting that two sets of hyperbolic constant-energy contours, which correspond to the splitting bands at $M_{\text{Ag(111)}}$, are observed in the bottom panel of Fig. 5(b) of Ref. [22] and Fig. S5 of the Supplemental Material [31]. This indicates that all the bands in the directions parallel to $\Gamma_{\text{Ag,Ge}} - \bar{K}_{\text{Ag,Ge}} - \bar{M}_{\text{Ag,Ge}}$ undergo umklapp-scattering effects with $G_{\text{Ag(111)}} = 2\Gamma_{\text{Ag(111)}}$. An intriguing question arises, “How can the Ag$_2$Ge surface alloy stabilize in spite of a $5\%$ lattice mismatch with the underlying Ag(111) surface?” Figure 4(a) shows the measured energy band dispersions of the Pb(111) dense phase in two symmetry directions, $\Gamma M$ and $\Gamma K$, after depositing extra Pb atoms onto the Ag$_2$Pb surface alloy. Note that green (red) symbols indicate the symmetry points of Pb(111) [Ag(111)], as shown from Fig. 4(c) that depict both SBZs. As was reported [34], the Ag$_2$Pb surface alloy undergoes a dealloying process at a coverage above 1/3 ML to form the Pb(111)-1 $\times$ 1 dense phase. In our experimental approach, we found that such a dealloying process can occur at RT without annealing. This implies the metastability of the Ag$_2$Pb surface alloy since the Pb atoms are only partially immersed in the alloy. Figure 4(d) shows the DFT calculation results on a one-layer freestanding Pb film in all major symmetry directions. The red curves superimposed onto the measured bands in Figs. 4(a) and 4(b) are those from Fig. 4(d) with a 0.6-eV offset. As seen, except for a $\Lambda$-shaped Ag bulk projected band edge centered at Ag(111) SZB, the bands with the orbital characters of $p_x$ and $p_y$ match the measured ones well but the bands of $p_z$ type do not have the counterparts in the measured spectra. The absence of $p_z$ bands in the measured spectra has occurred in several monatomic layers [3,6,35] and it was mainly ascribed to the interaction with the substrate due to the fact that their wave functions are out of the monolayer plane (See Supplemental Material Figs. S3(a) and S3(c) [31]). The same reason explains why the upward band centered at $\bar{\Gamma}$ for Ag$_2$Ge on Ag(111) revealed in the calculation, Fig. 3(c), is not observed in the measurement, Fig. 3(d), which is also a $p_z$ type (See Supplemental Material Fig. S4 [31] and Refs. 21,22). The calculated band structures in Fig. 4(d) show that the $p_z$ bands interact with the $p_x$, $p_y$ bands to cause a band gap merely above the Fermi level midways between $\Gamma$ and the SZBs, but Figs. 4(a) and 4(b) indicate the absence of such

FIG. 4. Energy band structures for 1-ML Pb(111) dense phase on Ag(111). (a), (b) The measured energy band dispersion, taken at 27-eV photon energy, of 1-ML Pb(111)-1 $\times$ 1 dense phase on Ag(111) in two symmetry directions with DFT calculated bands (red curves) superimposed from (d). (c) SBZs and symmetry points of Ag(111) (denoted in red) and 1-ML Pb(111) layer (denoted in green). $\bar{\Gamma}M_{\text{Ag(111)}} = 1.25$ Å$^{-1}$, $\bar{\Gamma}M_{\text{Pb(111)}} = 1.03$ Å$^{-1}$, $\bar{\Gamma}K_{\text{Pb(111)}} = 1.19$ Å$^{-1}$ (d) DFT calculated energy bands of one-layer freestanding Pb in symmetry directions.
band gaps because $p_z$ bands are pushed out of this energy range \( [9] \). Upon a further close examination, one can also see from Fig. 4(a) a band split at $M_{\text{Ag}(111)}$, which looks like a similar type (See Supplemental Material Fig. S2 \([31]\)) to that of Ag$_2$Ge on Ag(111), except that the splitting magnitude is larger and the more intense branch is at the left rather than at the right of $M_{\text{Ag}(111)}$. Therefore, the left belongs to the original Pb band and the right originates from the umklapp scattering mediated by Ag(111). The extracted splitting value, 0.214 Å$^{-1}$, between $M_{\text{Pb}(111)}$ and $M_{\text{Ag}(111)}$ corresponds to a lattice mismatch of 20.6%, close to the value of 20.5% derived from the bulk lattice constants of Pb and Ag, 4.92 and 4.08 Å. The LEED patterns in Fig. 5(a) exhibit 12 Moiré satellite spots around (00). It is actually made of 6 doublets. Each doublet, as enclosed by the yellow oval, spans ±21.5° with respect to the (00) point. This is the rotation angle of Moiré symmetry axis with respect to Ag(111). Figure 5(b) shows the same Moiré spots around the (10) points of Ag(111) in addition to the blurred residual spots of the Ag$_2$Pb layer at (1/3 1/3); however, the doublet, as enclosed by the yellow oval, spans ±4° with respect to (00). This indicates two domains of 1-ML Pb(111) films rotated ±4° with respect to the Ag(111)-1$\times$1 direction. Such a rotating behavior has been found in other film-substrate systems \([20,36,37]\) and is considered as a spontaneous mechanism for reducing large lattice mismatch. Taking into account the lattice constants of Pb(111) and Ag(111) as well as the ±4° splitting of LEED spots, the Moiré spots are reproduced via Eq. (1) as presented in Fig. 5(c). Note that the calculation for energy band dispersions in Fig. 4(d) does not consider the ±4° off the symmetry directions. The energy band dispersions close to those directions are shown in Supplemental Material Fig. S3(b) \([31]\), which show no relevant difference from those along symmetry directions.

Both large lattice-mismatched monatomic layers of Ag$_2$Ge alloy and 1-ML Pb film on Ag(111) show the common feature of the band splits at $M_{\text{Ag}(111)}$, generated by umklapp scattering via Ag(111). Such substrate-mediated umklapp electronic states in the context of incommensurate interface were observed as a second kind of quantum-well state (QWS) in Ag films \([38,39]\) or Pb films \([20]\) at a thickness above 1 ML on Ge(111). The discrete momentum perpendicular to the surface, $k_{\perp}$, of QWS has to be taken into account for the phase accumulation model although it is the momentum parallel to the surface, $k_{||}$, which induces umklapp scattering.

For the case of a monatomic layer, there is no $k_{\perp}$ of the electronic states so the corresponding energy band structures of the monolayer electrons undergoing the substrate-mediated umklapp effect would be simply offset by the reciprocal vector of the substrate surface, as clearly illustrated in Fig. 2(e). It is worth mentioning that the crossing points of the splitting bands do not reveal relevant band gaps for the Ag$_2$Ge alloy and 1-ML Pb film on Ag(111) possibly due to a weak super-lattice potential \([40]\). Because the second kind of QWS, as the first kind, partially contribute to the total surface energy of a film, the monolayer-umklapp-electron states likely do so for a monolayer on an incommensurate interface, and are hence related to the monolayer stability. In addition, although an incommensurate interface is not appropriate for the optimal bonding between atoms of films and substrates to substantially reduce energies, partial bonding can still be achieved as evidenced by the absence of the $p_z$-type band in the measured energy bands of the Ag$_2$Ge layer and 1-ML Pb on Ag(111).

**IV. CONCLUSION**

In conclusion, comparing the two systems, Ge and Pb on Ag(111) at the coverage 1/3 ML, a long-time overlooked factor was discovered that the sizes of adsorbate atoms relative to the substrate atoms are relevant to the lattice and electronic structures of the initial alloy layer and likely further affect subsequent overlayers. For Ag$_2$Pb on Ag(111), the tensile strain induced by the larger Pb-atom size is released by the corrugation to maintain a Ag$\sqrt{3}$-R30 commensurate lattice, while for Ag$_2$Ge on Ag(111), the compressive strain due to the smaller size of the Ge atom leads to $\sim -5\%$ lattice mismatch at the interface where umklapp scattering mediated by Ag(111) causes the observed band split at $M_{\text{Ag}(111)}$. This was further confirmed by the similar behavior observed from 1-ML Pb layer on Ag(111). We solve the puzzle of apparent Rashba band splitting at the substrate SZB for the Ag$_2$Ge surface alloy on Ag(111) in light of the incommensurate interface. The relation among the composite-atom sizes, the strain percentage with respect to Ag(111), and the band-splitting magnitude can be systematically studied by including other Ag$_2$M families (e.g., $M = \text{Sn}, \text{Bi}, \text{As}$, etc.) for a completely quantitative comparison (See Supplemental Material Table S1 \([31]\)).
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