Significantly enhanced giant Rashba splitting in a thin film of binary alloy

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Supplementary Figures:
Supplementary Figure 1: A series of LEED patterns for the incremental deposition of Au onto the 6-ML Pb films with their corresponding photoemission spectra. As seen, when 0.4 Å Au is deposited, the single incommensurate PbAu alloy layer forms on top of the Pb film, and two Dirac cones are not yet observed in the corresponding photoemission spectra. When the deposited Au increases to 1.6 Å, the spots originating from the underlying Pb films vanish and the two Dirac cones start clearly observed. At this stage, Kagome capping Au layer forms on top of the PbAu alloy layer. With further deposition of Au, the LEED patterns maintains the same but the intensity of two Dirac cones start decreasing at 4.8 Å of Au deposition. All these evolutions indicate that the two Dirac cones are most pronouncedly observed at about 2 Å of Au deposition, at the stage of which one layer of capping Au forms on top of the PbAu alloy layer.

Supplementary Figure 2: The calculated band dispersions of the single-freestanding PbAu alloy layer in a relaxed form depicted in Figure 5(c). Each band shown is double degenerate, indicating no Rashba effect at all. The energy offset of Fermi level is not adjusted because no corresponding data can be compared with.
Supplementary Figure 3: The spin-orbital-splitting dependence of the calculated band dispersions of the PbAu alloy in $\Gamma - \bar{M} - \Gamma$ direction with a special buckling configuration induced by the squeezing between the top Au layer and bottom Pb layers. The determination of Rashba energy $E_R$ and momentum offset $K_0$ are extracted according to this evolution.

Supplementary Figure 4: The spin-orbital-splitting dependence of the calculated band dispersions of the PbAu alloy in $\bar{M} - \bar{K} - \bar{M} - \bar{K}$ direction with a special buckling configuration induced by the squeezing between the top Au layer and bottom Pb layers. Supplementary Figure 4(a) corresponds to Figure 5(b) in the paper. As seen, the energy difference between the top Rashba splitting (TR) and bottom Rashba splitting (BR) at $\bar{M}$ as well as the momentum offsets of both splitting increase with SO strength.
Supplementary Figure 5: The buckling-height dependence of the calculated band dispersions of the PbAu alloy in $\overline{\Gamma}-\overline{K}-\overline{M}-\overline{K}$ direction. Supplementary Figure 5(a) corresponds to Figure 5(b) in the paper. As seen, the energy difference between the top Rashba splitting (TR) and bottom Rashba splitting (BR) at $\overline{M}$ as well as the momentum offsets of both splitting increase with buckling height.
Supplementary Figure 6: Photon energy dependence of the top cone and bottom cone of the PbAu alloy, indicating that they are 2D electronic structures.
Supplementary Figure 7: Photoemission data for the PbAu alloy covered with one layer of Au on 2-ML Pb film /Ge(111). (a) Energy band dispersions in symmetry directions $\Gamma - K - \bar{M}$ and (b) $\Gamma - \bar{M} - \Gamma$. (c) The corresponding LEED pattern.
Supplementary Figure 8: Photoemission data for the energy band dispersions of (a) 6-ML Pb film and (b) Pb(111) bulk crystal in the same symmetry direction $\Gamma - \bar{M}$. 