Supplementary material for:
"Rare-earth surface alloying: a new phase for $GdAu_2$"
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ARPES DETAILS

E-E$_f$($\overline{\Gamma}$) $\pm$ 0.05 eV	SS	D	E	F	A	B	G
0.5 ML $GdAu_2$	-0.45						
1.0 ML $GdAu_2$					-1.05	-1.80	
2.0 ML $GdAu_2$		-0.25	-0.40	-0.70	-1.05	-1.80	-2.25

TABLE I: Experimental values in (eV) for the binding energies belonging to the different bands identified in Figure 1 and 2 of the main article. All the values are given with respect to the Fermi level (E_F) at $\overline{\Gamma}$ and are estimated with $\pm$0.05 eV as error bar.

Angle Resolved Photoemission experiments (ARPES) were carried out using synchrotron light from the PGM beamline of the Synchrotron Radiation Center (SRC) in Stoughton (Wisconsin). We used a hemispherical Scienta SES200 spectrometer with energy and angular resolution set to 25 meV and 0.1°, respectively, and p-polarized light. Measurements were taken at 150 K.

Table I shows the band positions for Fig. 1 and Fig. 2 of the main paper.

FERMI SURFACE CHARACTERIZATION

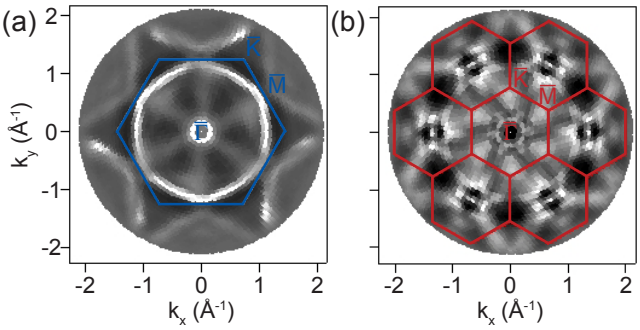

FIG. 1: Fermi surface maps of (a) clean Au(111) and (b) 2 ML $GdAu_2$/Au(111). Data were recorded with HeIα radiation (21.2 eV). The 1 × 1 corresponding surface Brillouin zones and high symmetry points $\overline{\Gamma}$, $\overline{K}$ and $\overline{M}$ are overlayed.

Figure 1 shows the Fermi surface maps measured with HeIα radiation for clean Au(111) (a) and 2 ML $GdAu_2$/Au(111) surface compound (b). The 1 × 1 corresponding surface Brillouin zones (SBZ) are superimposed. The compound SBZ is rotated 30° with respect to the substrate one, as already found in Low Energy Electron Diffraction patterns (LEED). It is defined by the reciprocal lattice vectors $d_{\overline{\Gamma K}}$ = 0.77 Å $d_{\overline{\Gamma M}}$ =0.67 Å which coincide with a distance of d= 5.43 Å between equivalent atomic species in the direct space. The values for the workfunctions measured for the clean and alloyed surface are: 4.52 eV and 4.72 eV respectively.

X-RAY DIFFRACTION AND SIMULATIONS

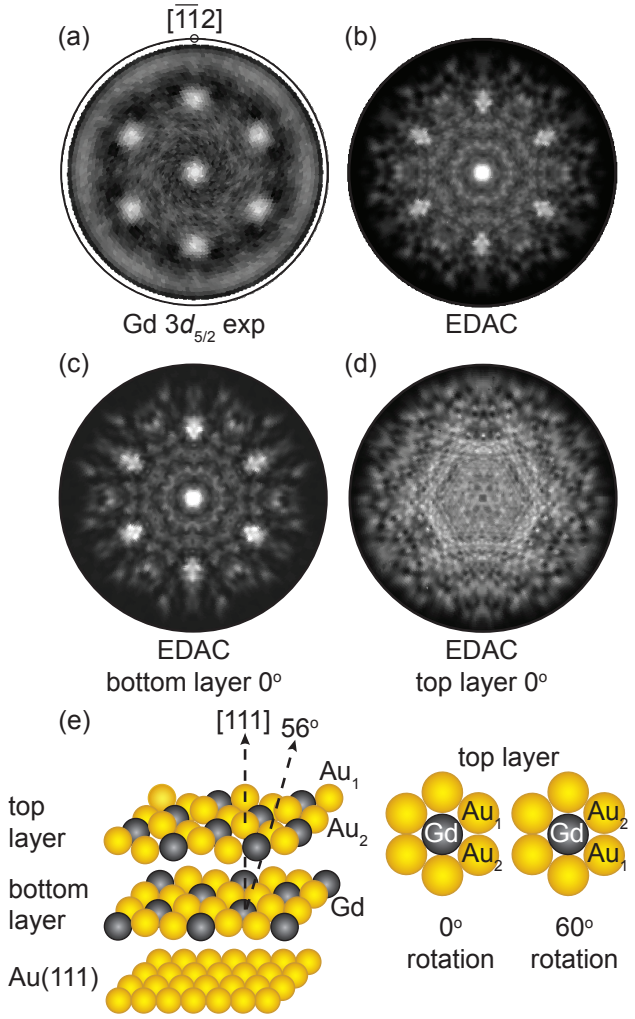

FIG. 2: (a) Experimental X-ray photoelectron diffraction pattern recorded at the Gd 3$d_{5/2}$ emission (E$_{kin}$=553 eV) for the 2 ML $GdAu_2$/Au(111) compound. (b-d) Multiple scattering calculation (MSC) for the pattern in (a) simulated with the EDAC code [1]. The total pattern in (b) results from the sum of 4 MSC simulations for Gd atoms in the top (d) and bottom (c) $GdAu_2$ layers when the top layer is rotated 0° (for c and d) or 60°(not shown) degrees with respect to the bottom one.

Crystallographic directions and orientation are given by the substrate Au 4$f_{7/2}$ XPD patterns, recorded simultaneously with the Gd 3$d_{5/2}$. The Au 4$f_{7/2}$ (E$_{kin}$=1656

eV) diffractogram (not shown) is dominated by bulk emission, so that features from the surface Au atoms are greatly depleted.

The atomic cluster employed for the simulations consists of two shifted alloy layers and a flat Au 1×1 unreconstructed substrate, with 100 atoms in a parabolic surface focused on the emitter Gd. Figure 2(c) shows the theoretical pattern calculated with the EDAC code [1] for a Gd emitter placed in the bottom $GdAu_2$ layer that, as compared to Fig. 2(a), is the main responsible for the intensity distribution in the experimental measurement. Fig. 2(d) is the simulated diffraction pattern generated by a Gd atom in the top $GdAu_2$ layer. Both Fig. 2(c) and (d) show a 3-fold symmetry due to photoelectron diffraction with the highest equivalent Au atoms on the surface. They correspond to a top $GdAu_2$ layer not rotated (0°). Equivalent patterns (here not shown) are calculated for the other possible atomic arrangement with the top layer 60° rotated to the bottom one. The sum of the four patterns is displayed in Fig. 3(b) of the main article. The possibility to obtain a single domain has not been investigated so far.

DETAILS OF THE THEORETICAL CALCULATIONS

The electronic structure calculations are performed using the ABINIT package[2, 3], in the local density approximation and the projector augmented-wave[4] formalism (plane wave cut off at 20 Ha). The Gd potential is generated with 18 electrons in the valence (2.54 a.u. cutoff radius) whereas the Au potential has 11 electrons in valence (with a 2.69 a.u. cutoff). Both have 2 projectors per l channel. The Brillouin Zone sampling of 8×8 is effectively 16×16 for the underlying gold unit cell, ensuring good convergence for the extended bulk Au states. The slabs used are 7ML thick. The overlayers and the first two surface layers of gold are fully relaxed until either the forces were smaller than 5×10^{-5} Ha/bohr, or displacements smaller than 1 pm. The in-plane lattice constant is fixed to the theoretical relaxed value for bulk Au. The spin-orbit interaction is included in all calculations. Further, as the *f*-electron correlation is quite strong, we employ the LDA+U scheme [5] (U=6.7 eV J=0.7 eV) in the Fully Localized Limit, and applied only on the f electrons of the Gd. This pushes the *f* bands down to -9 eV.

Bulk tetragonal $GdAu_2$ is antiferromagnetic between planes[6], and this is correctly preferred by our ab-initio calculations. The AFM order also improves the bulk lattice constant from 15% to less than 1% error with respect to experiment.

For the STM calculations, the correspondence of currents in absolute values is not possible due to the approximations and lack of atomic information about the tip structure.

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