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Supporting Information

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Refined Epitaxial Growth of $YbRh_2Si_2$ Thin Films

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Supporting Information

Table S1: Elemental compositions [%] according to energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy (XPS). The XPS spectra to extract the composition are acquired after sputtering the samples for 5 minutes.

Sample	Yb EDX Composition [%]	Rh EDX Composition [%]	Si EDX Composition [%]	Yb XPS Composition [%]	Rh XPS Composition [%]	Si XPS Composition [%]
A	18.2	42.4	39.4	19.2	51.4	29.4
B	19.0	41.4	39.6	20.6	51.4	28.0
C	19.5	41.1	39.4	19.9	50.2	29.9
D	19.1	42.2	38.7	20.0	51.3	28.7
E	20.0	42.8	37.2	19.5	52.8	27.7
F	19.2	41.4	39.3	19.9	51.1	29.0
G	20.2	42.0	37.8	20.9	51.4	27.7
H	20.1	40.6	39.3	20.7	50.3	29.0

Energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy (XPS) measurements were performed to extract Yb, Rh, and Si compositions. A Physical Electronics PHI Versaprobe III with a hemispherical energy analyzer and a monochromatic aluminum Kα X-ray source (1486.6 eV) was used for XPS. The instrument's charge neutralization system prevented sample charging. Data were acquired using a 200 µm, 50 W focused X-ray beam at a base pressure of 1×10^{-9} mbar, and a take-off angle of 45°. The survey scans were collected with a pass energy of 140.00 eV and a step size of 0.5 eV. High-resolution scans of peaks of interest were collected with a pass energy of 27.00 eV and a step size of 0.05 eV. Data were analyzed with CASA XPS software. Spectra of the pristine sample, as well as after sputtering the samples for 5 minutes using Ar^+ ion beam with an energy of 1 keV and an incidence angle of 60° were acquired.

The measured elemental compositions of the samples are in good agreement with EDX, as reported in Table S1. The XPS spectra obtained from sample F are shown in Figure S1, before and after sputtering. As can be observed, the concentration of rhodium is systematically higher than what is expected from the beam calibration and from what was measured by EDX, while the concentration of silicon is always lower than the reference. This can be explained by the fact that rhodium is naturally resistant to oxidation. The samples formed an oxide on the surface that is mainly constituted of silicon and with a minor contribution of ytterbium. The spectrum of the pristine sample shows a clear peak for oxidized silicon at around 102 eV. Rh has only a concentration of 10% in the pristine sample. To perform a correct quantification through XPS, the sample were sputtered for 5 minutes using Ar^+ ions. This process has two consequences: First, it removes the surface oxide, which has the consequence of increasing the relative concentration of Rh in the sputtered sample. Secondly, the sputtering process can remove different elements at a different rate, which explains the observed lower concentration of Si.

Figure S2 shows the X-ray diffraction (XRD) $\omega - 2\theta$ scan of the epitaxial $YbRh_2Si_2$ films grown on Ge(0 0 1) at different Yb fluxes.

Figure S3 shows the in-plane lattice parameter a of the epitaxial $YbRh_2Si_2$ films grown on Ge(0 0 1) as a function of the Yb, Rh, and Si composition. The values of a are extracted from RHEED, the Yb, Rh, and Si elemental composition are extracted from EDX analysis.

Figure S4 shows the side and top view of the optimized structure of the Ge slab model used for the density functional theory (DFT) calculations. The two bottom layers are fixed, and a vacuum layer is added to the top to avoid spurious interactions with neighboring slabs from periodic boundary conditions.

Table S2 summarizes the parameters and results of the epitaxy of $YbRh_2Si_2$ films after soaking the surface of the Ge substrates with 0 to 1 ML of Yb.

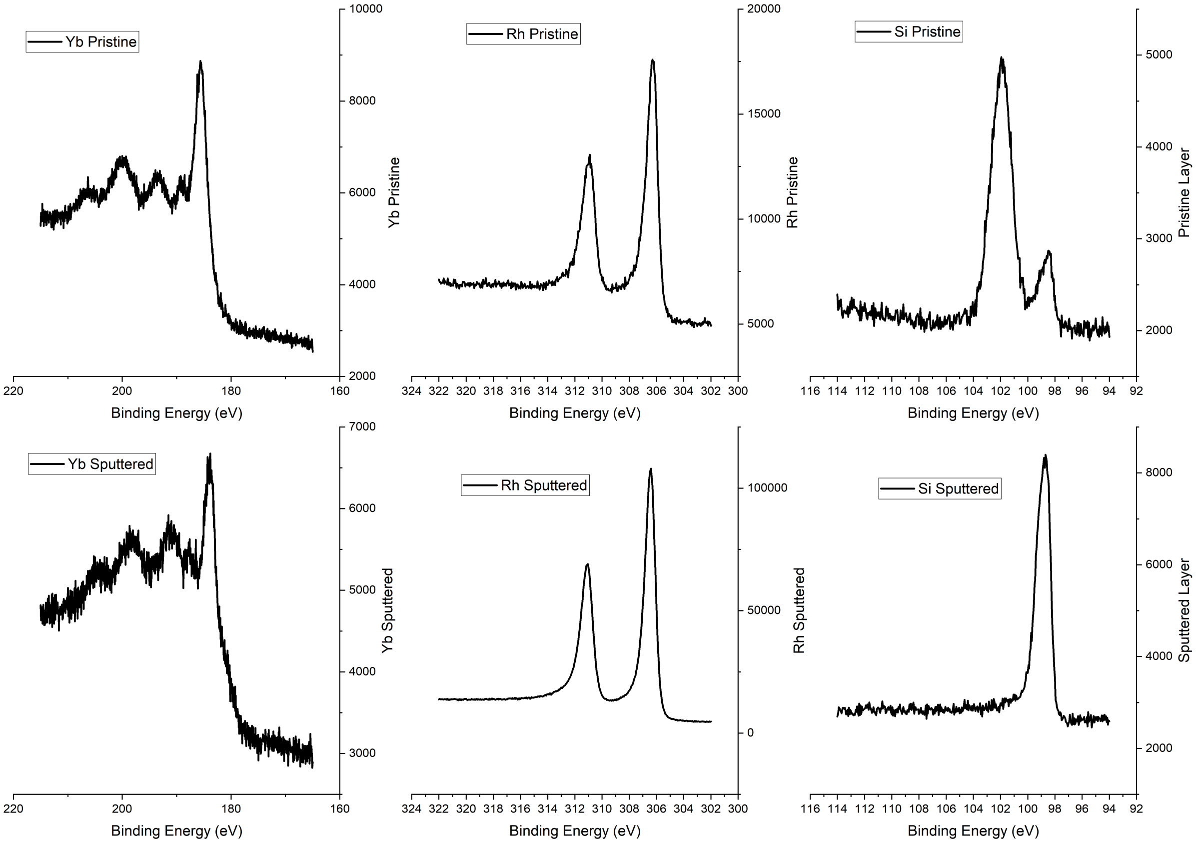

Figure S1: XPS spectra of Yb, Rh and Si before and after 5 minutes of sputtering of sample F.

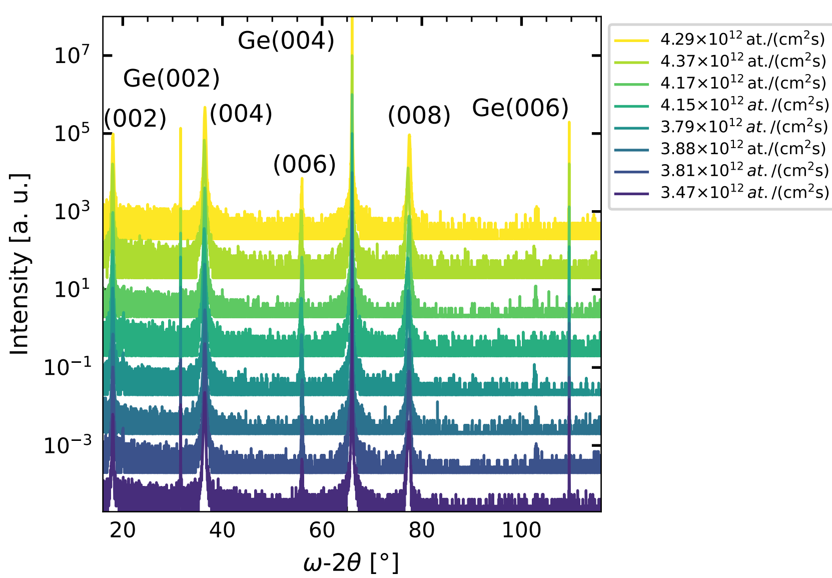

Figure S2: X-ray diffraction (XRD) $\omega - 2\theta$ scan of the epitaxial $YbRh_2Si_2$ films grown on Ge(0 0 1) with different Yb fluxes. The curves are plotted on a logarithmic scale and vertically shifted for clarity.

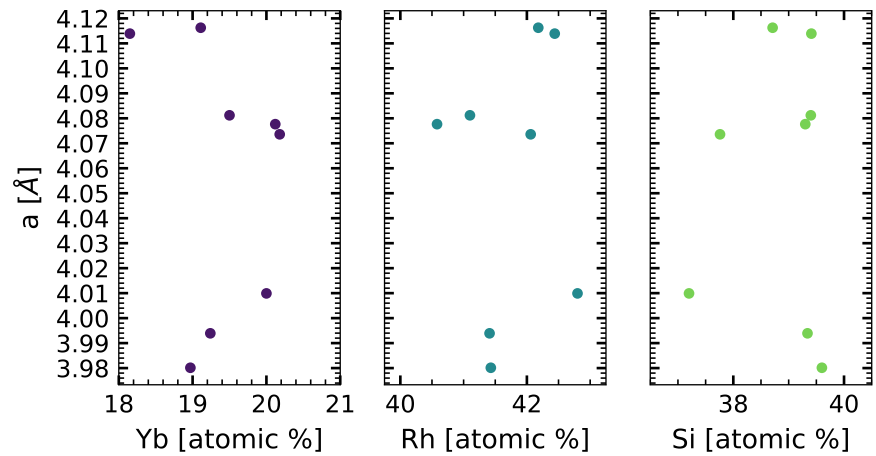

Figure S3: Lattice parameter a of the epitaxial $YbRh_2Si_2$ films grown on Ge(0 0 1) as a function of the Yb, Rh, and Si composition. The values of a are extracted from reflection high-energy electron diffraction (RHEED).

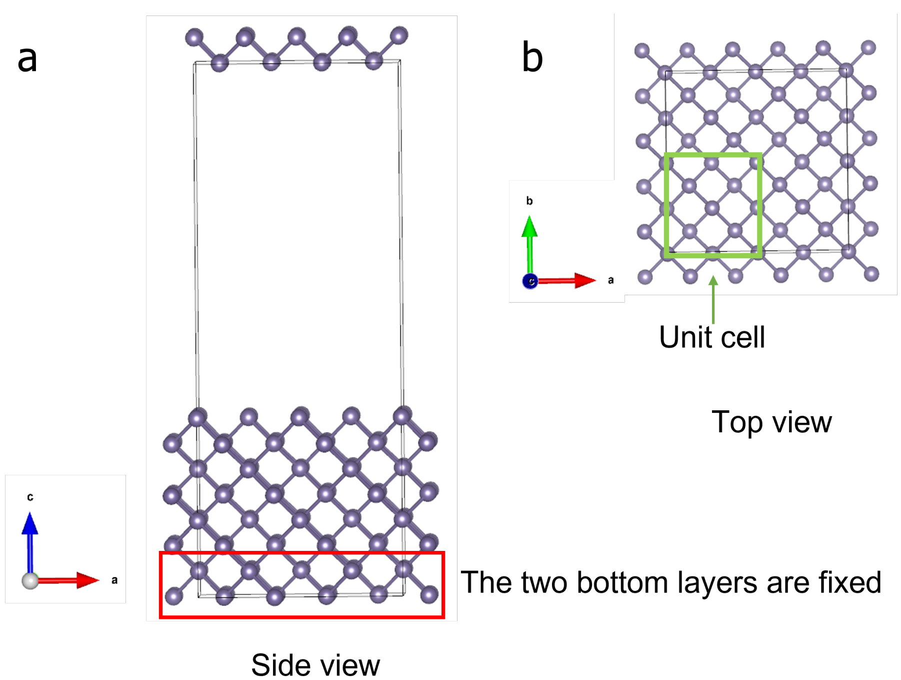

Figure S4: Side (a) and top (b) view of the optimized structure of the Ge slab model.

Table S2: Growth temperature, soaking element, number of MLs, and AFM RMS roughness of 10-nm-thick $YbRh_2Si_2$ films.

Growth temperature [°C]	Soaking element	Number of monolayers (MLs)	Atomic force microscopy (AFM) root mean squared (RMS) roughness [nm]	AFM skewness
400	-	0	0.692	-1.542
400	Yb	1/3	1.840	-7.814
400	Yb	2/3	1.118	-0.185
400	Yb	1	0.941	-4.527
425	-	0	3.224	-0.950
425	Yb	1/3	4.430	-1.041
425	Yb	2/3	1.704	-0.664
425	Yb	1	1.916	-1.209
450	-	0	4.818	-1.294
450	Yb	1/3	3.560	-1.523
450	Yb	2/3	3.227	-1.996
450	Yb	1	2.944	-0.600
475	-	0	4.990	-1.156
475	Yb	1/3	6.741	-0.247
475	Yb	2/3	3.900	-0.514
475	Yb	1	1.816	-0.423