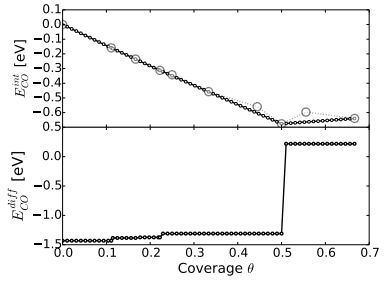
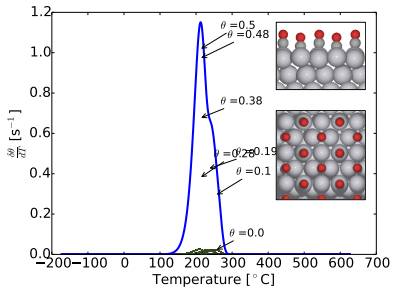
A Supplementary Material

Adsorption energies and simulated TPD spectra are shown for surfaces with strain in the range from −6% to 0%. Each TPD spectrum is the sum of spectra at every coverage θ from 0 to 0.5 ML resolved in 65 sites. Differential adsorption energies are calculated by differentiating the interpolated convex hull of the integral adsorption energy. The vibrational entropy contribution was TS_{vib} = 0.139 eV and the zero-point energy was 0.21 eV for CO adsorbed on top at 0% strain and the ZPE was 0.17 eV for CO in vacuum. The relevant reference state is CO desorbing in UHV, and thus the entropy contribution is quite uncertain. We have chosen to neglect the entropy of the reference state. Thus, a constant value of -0.1 eV was used for $\Delta ZPE - T\Delta S$ at all strains and coverages.

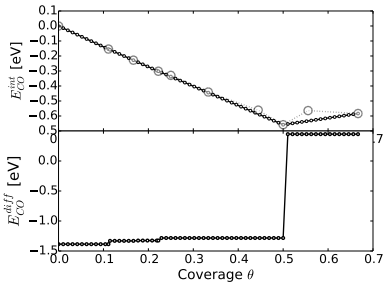


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

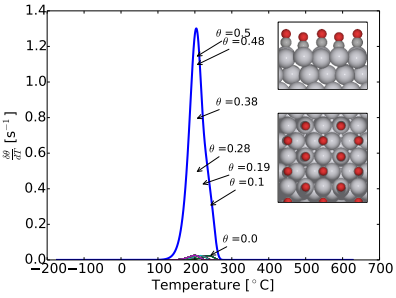


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.1: *A relaxed Pt(111) surface.*

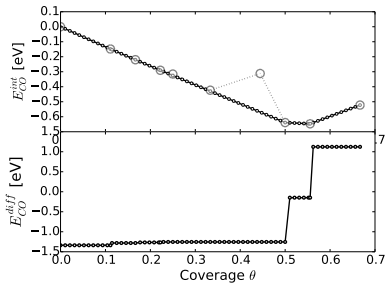


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

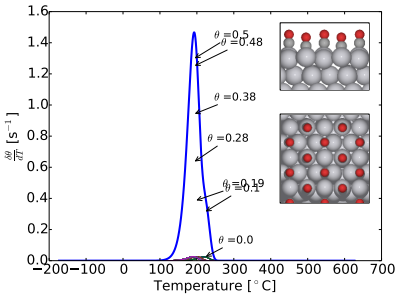


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.2: *Pt(111) surface under 1% compressive strain.*

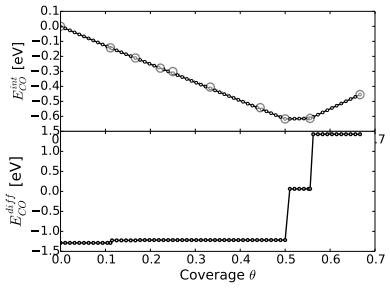


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

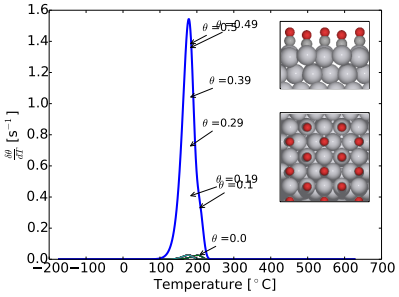


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.3: *Pt(111) surface under 2% compressive strain.*

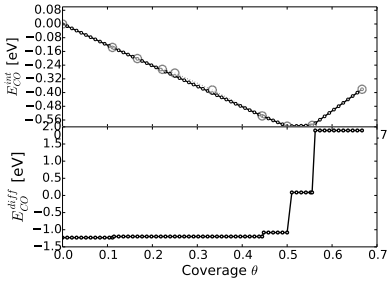


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

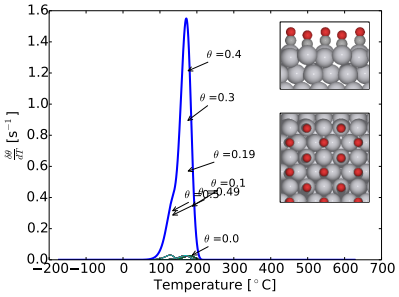


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.4: *Pt(111) surface under 3% compressive strain.*

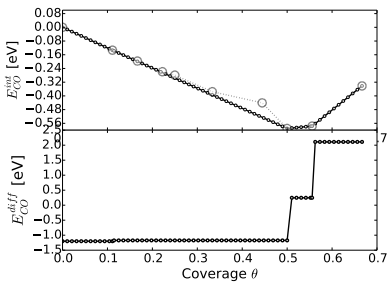


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

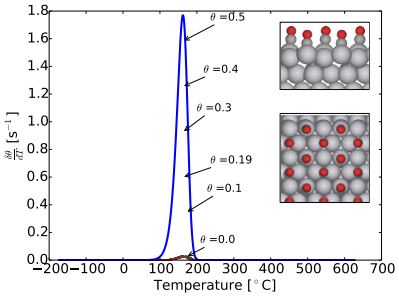


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.5: *Pt(111) surface under 4% compressive strain.*

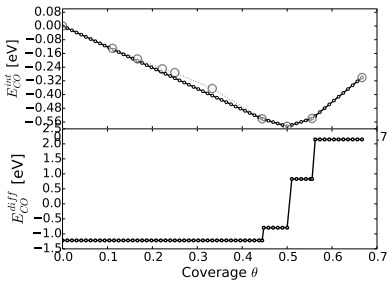


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

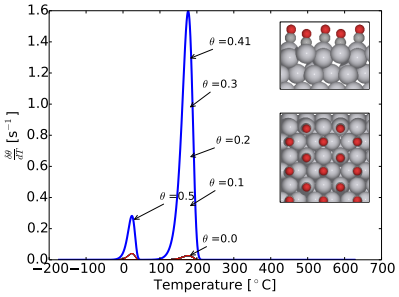


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.6: *Pt(111) surface under 4.5% compressive strain.*

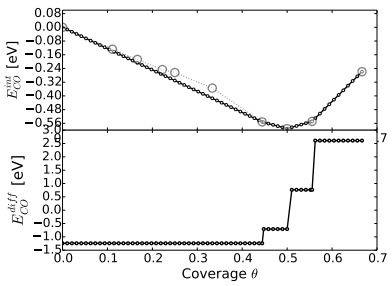


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

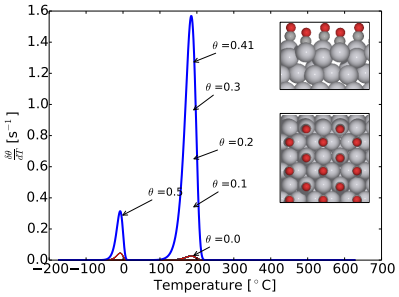


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.7: *Pt(111) surface under 5% compressive strain.*

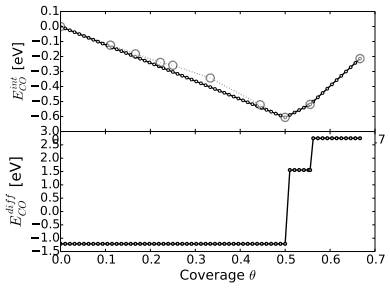


(a) *Energies of adsorption. Upper panel shows* ΔE^{int} *of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*

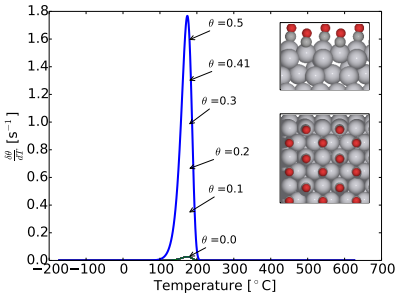


(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each* $\Delta E^{diff}(\theta)$ *are shown as thin lines.*

Figure A.8: *Pt(111) surface under 5.5% compressive strain.*



(a) *Energies of adsorption. Upper panel shows ΔE^{int} of DFT calculations as large symbols and interpolation of the convex hull as small symbols. Lower panel shows the differential adsorption energy.*



(b) *Simulated TPD curves. The blue line is the sum of the spectra calculated from individual differential energies at all coverages. Individual spectra from each $\Delta E^{diff}(\theta)$ are shown as thin lines.*

Figure A.9: *Pt(111) surface under 6% compressive strain.*

B XPS measurements

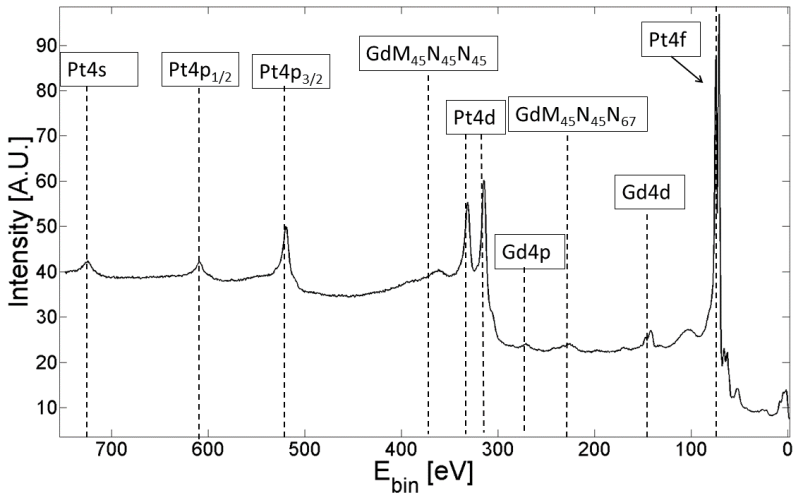


Figure B.1: *XPS measurement made on the prepared Gd/Pt(111) surface using a Mg anode.*

XPS was made to verify the cleanliness of the prepared Gd/Pt(111) sample. These experiments did not show any detectable traces of C or O, even after 20 cycles. An XPS overview measurement can be seen in B.1. The 4f peak from Pt and the 4 d peak from Gd was used to evaluate the composition. The stoichiometry of the alloy has been evaluated assuming that the platinum skin consists of a single layer of atomic platinum of thickness $d = 2.28$ Å [Briggs and Seah(1990)], see Table B.1.

Pt [%]	Gd [%]	x for Pt/Pt$_x$Gd
87.4	12.6	5.5

Table B.1: *Concentrations of the different elements in the Gd/Pt(111) quantified with XPS after the gadolinium deposition.*

C Ion Scattering Spectroscopy measurement

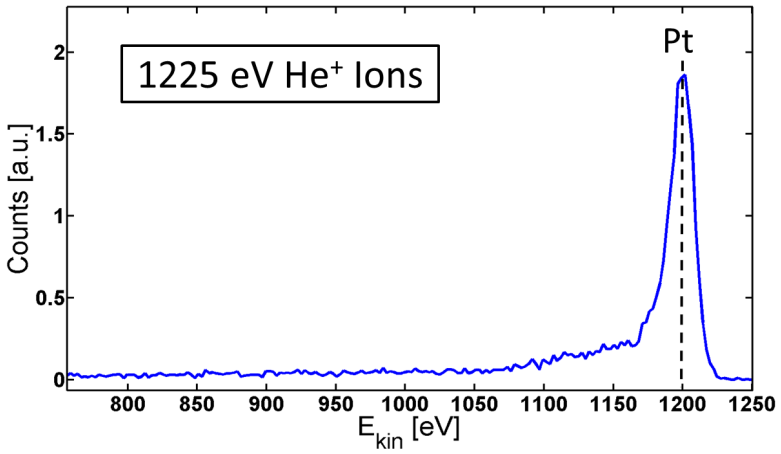


Figure C.1: *ISS measurement made on the prepared Gd/Pt(111) surface using a* He^+.

The composition of the outermost surface layer was evaluated using ISS, see Figure C.1. This experiment showed that the surface was terminated by a layer of Pt.

D LEED images after CO dosage.

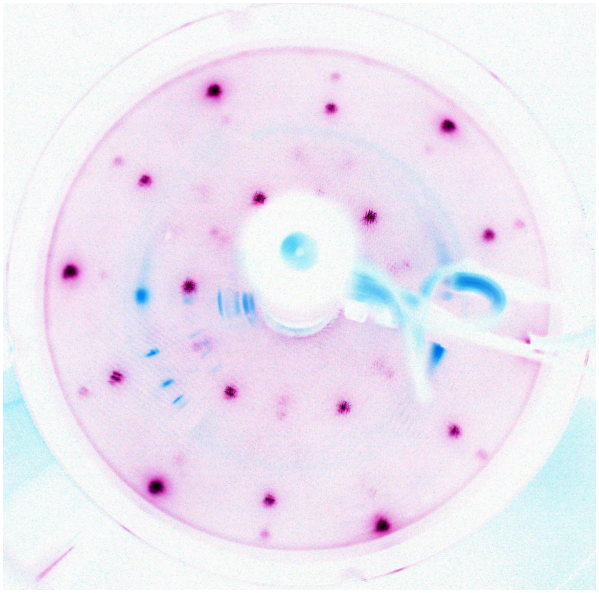


Figure D.1: *LEED was made after 1 L of CO had been dosed.*

References

[Briggs and Seah(1990)] D. Briggs and P. Seah, *Practical Surface Analysis, Auger and X-ray Photoelectron Spectroscopy*, Wiley, 1990.