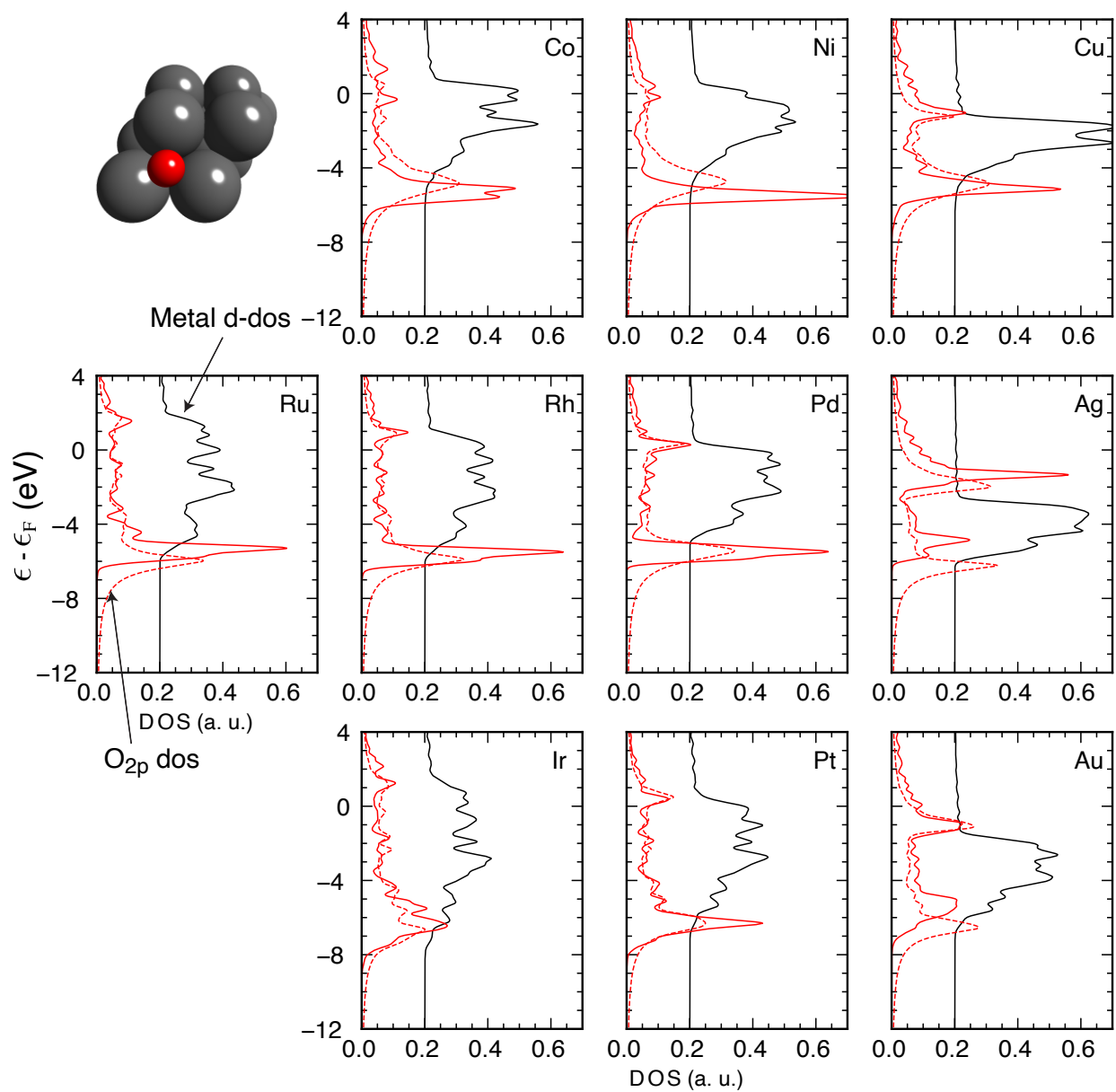
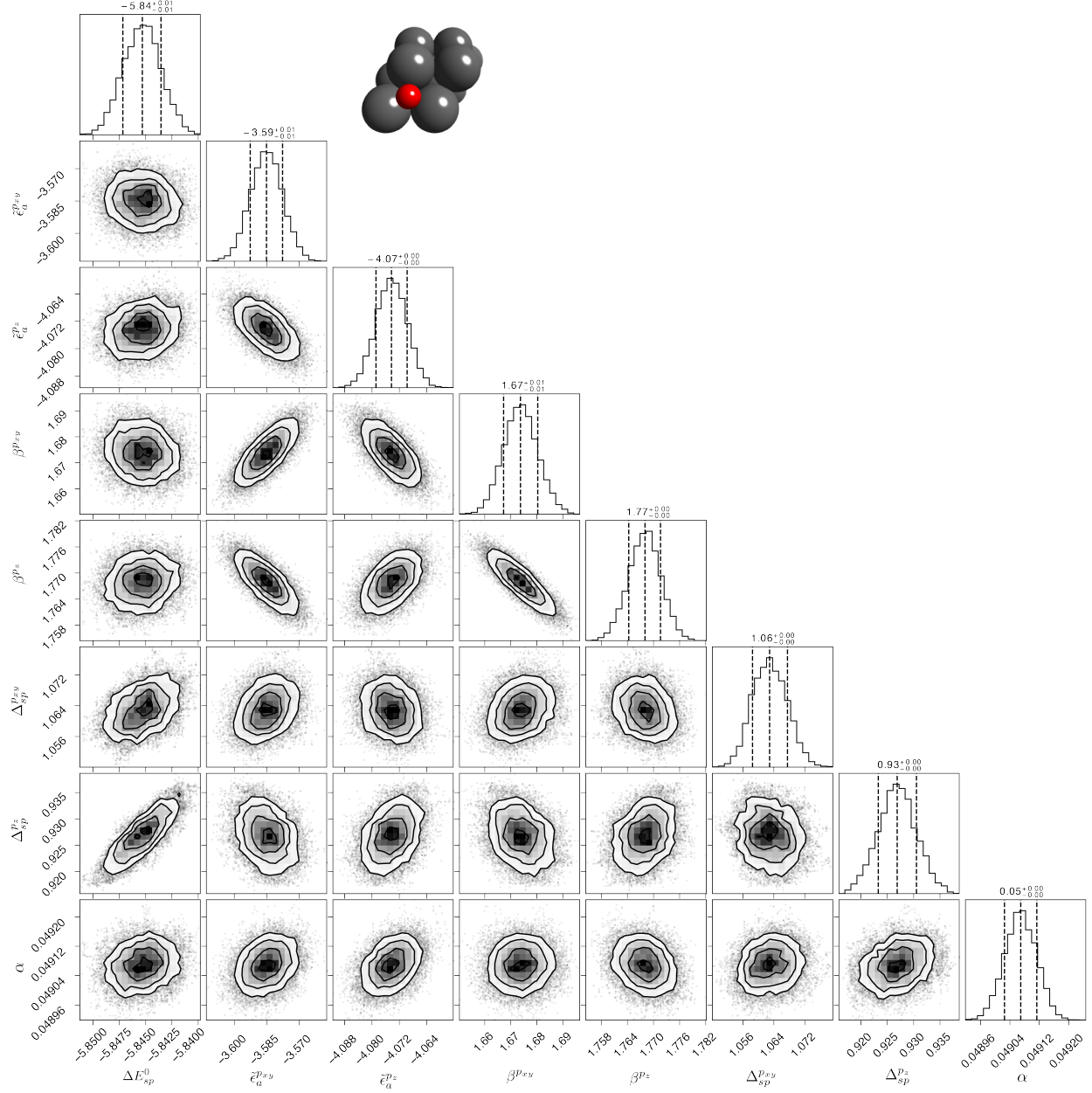


Supplementary Materials: Bayesian Learning of Chemisorption for Bridging the Complexity of Electronic Descriptors

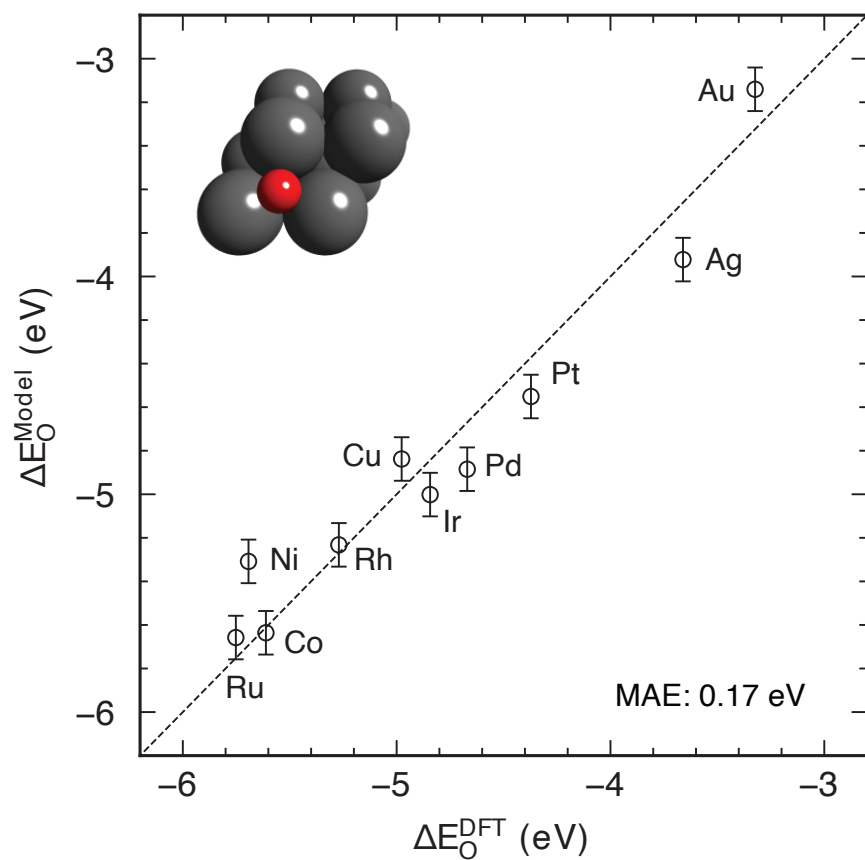
Siwen Wang, Hemanth Somarajan Pillai, Hongliang Xin



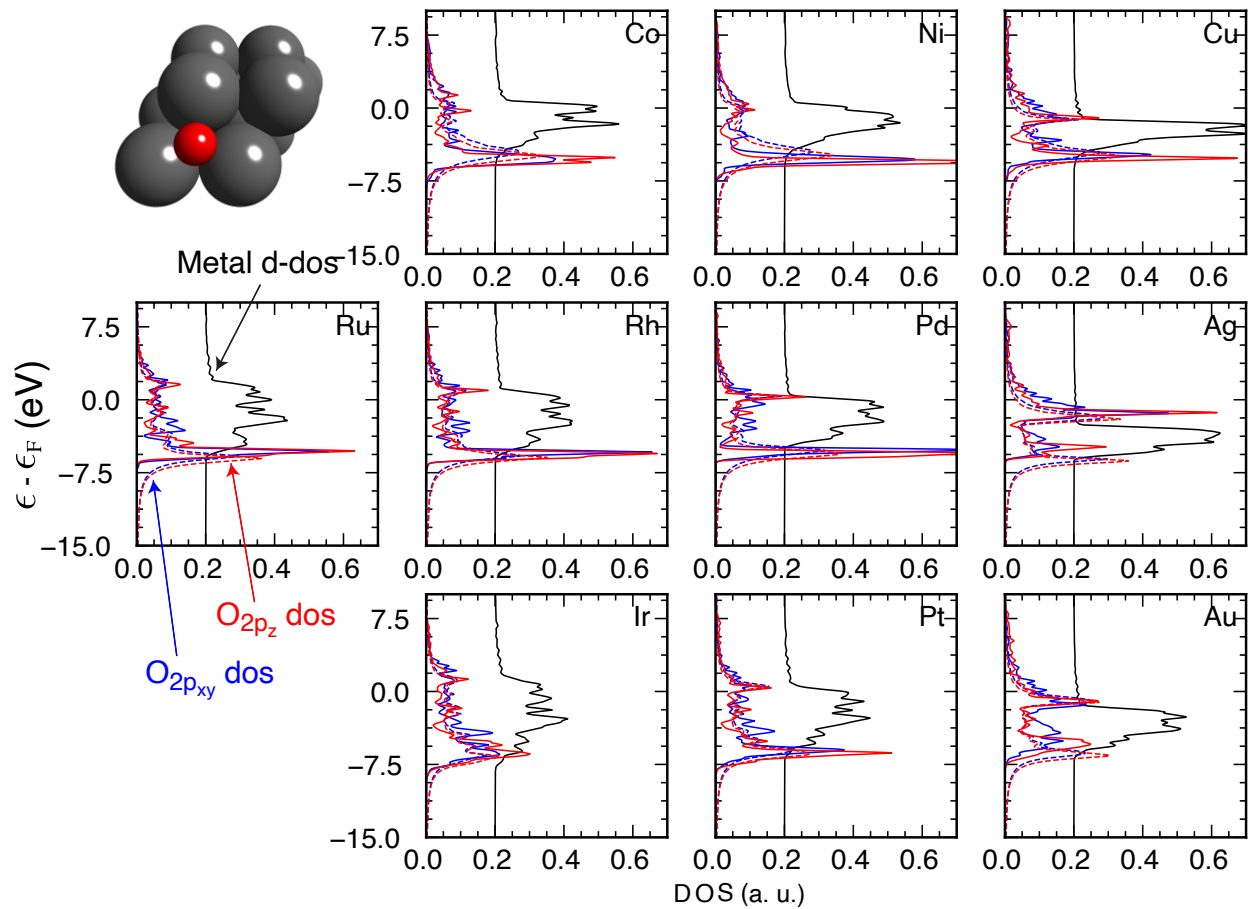
Supplementary Figure 1. Projected O_{2p} density of states from DFT calculations (solid) and model prediction (dashed) using the Bayesian-learned posterior means of single-orbital model parameters. Metal d -states are also shown.



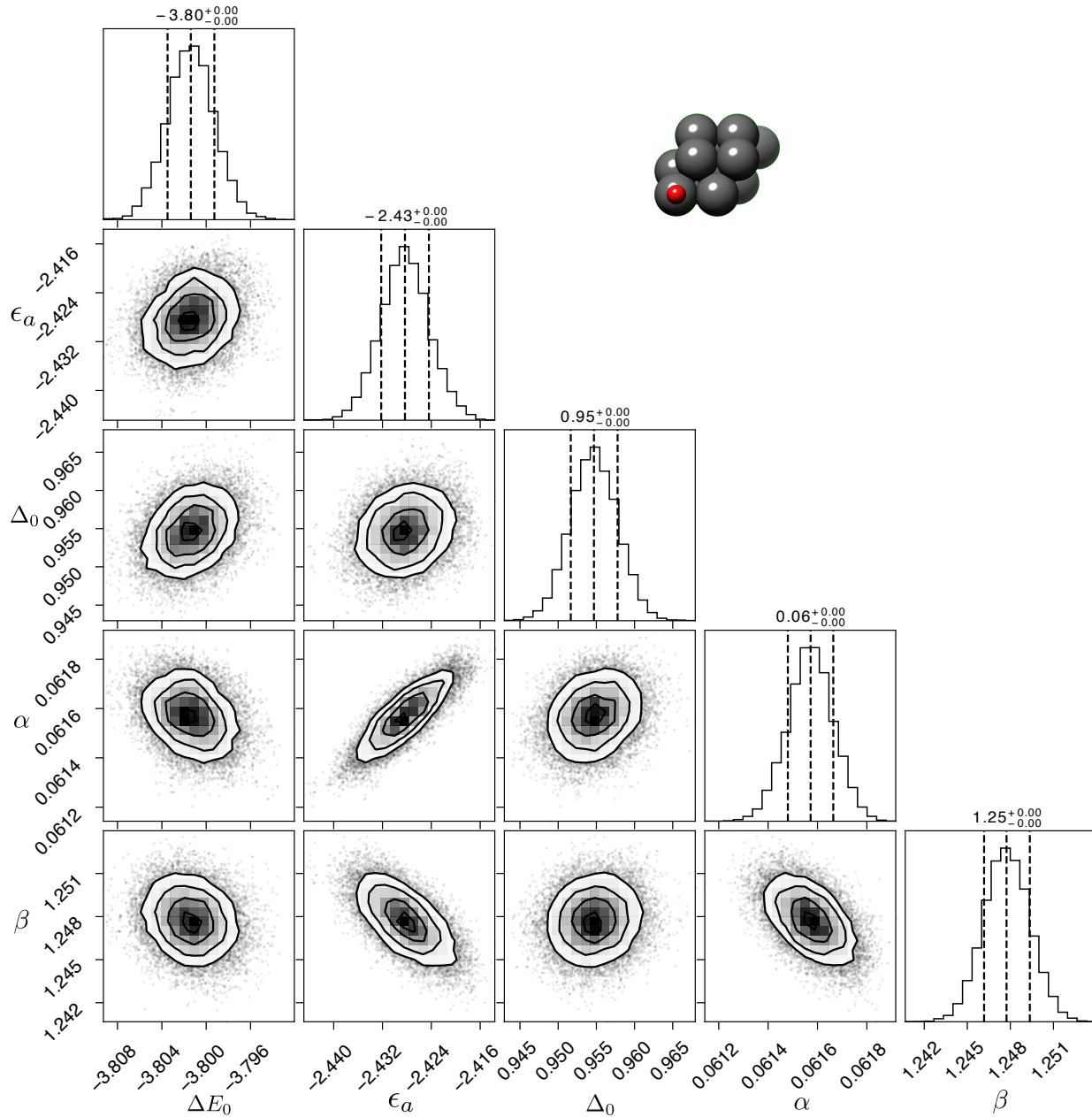
Supplementary Figure 2. The co-variance of the joint posterior distribution for each parameter pair and 1D histogram of multi-orbital model parameters (ΔE_0 , $\epsilon_a^{p_{xy}}$, $\epsilon_a^{p_z}$, $\beta^{p_{xy}}$, β^{p_z} , $\Delta_0^{p_{xy}}$, $\Delta_0^{p_z}$, and α) from MCMC simulations for *O adsorption at the fcc-hollow site of the close-packed, pristine transition-metal surfaces.



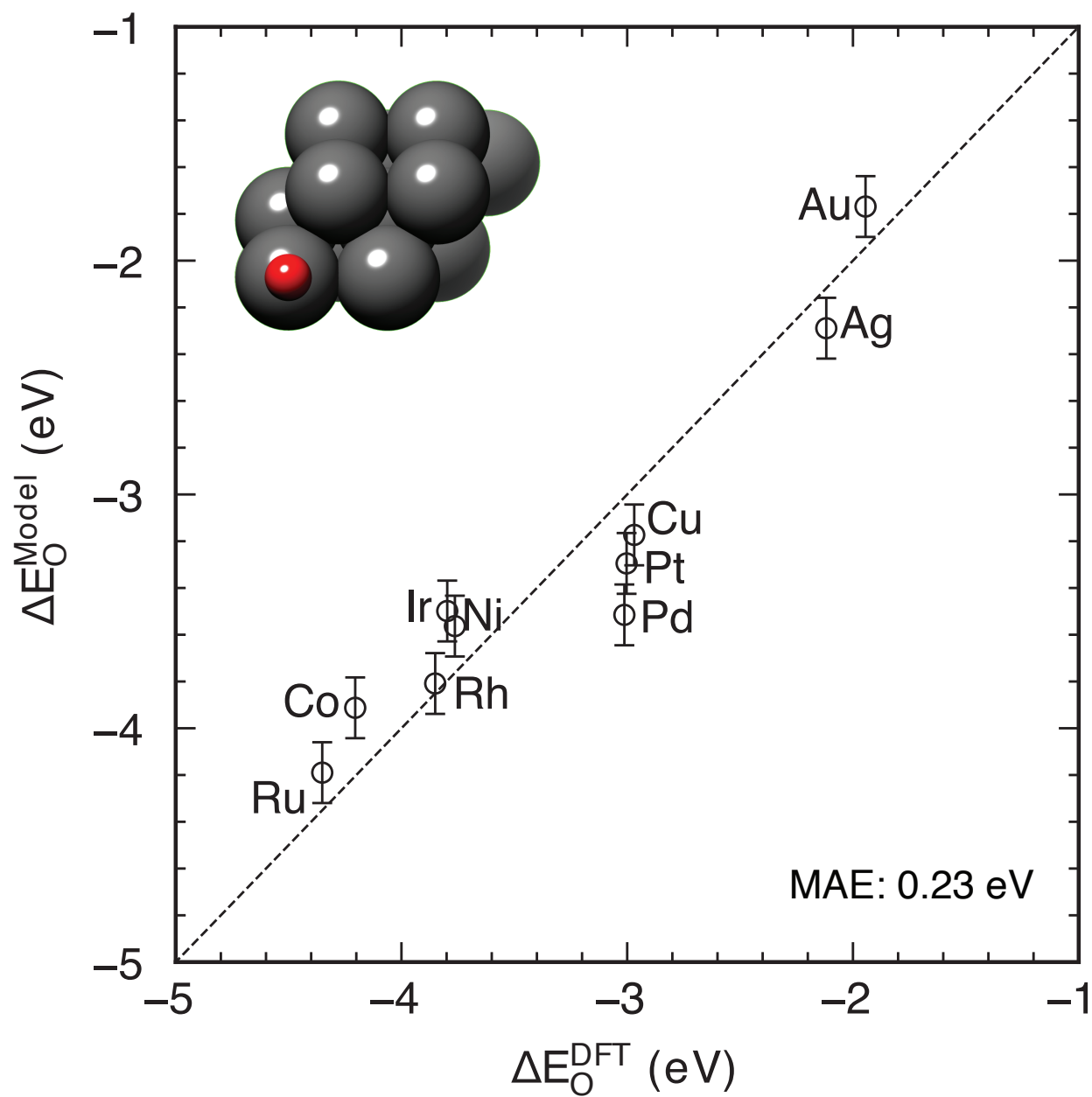
Supplementary Figure 3. DFT-calculated $\ast\text{O}$ adsorption energies at metal surfaces vs. model prediction using the posterior distribution of multi-orbital model parameters ($\vec{\theta}$, $\vec{\sigma}$). Error bars represent the standard deviation of prediction based on Bayesian sampling.



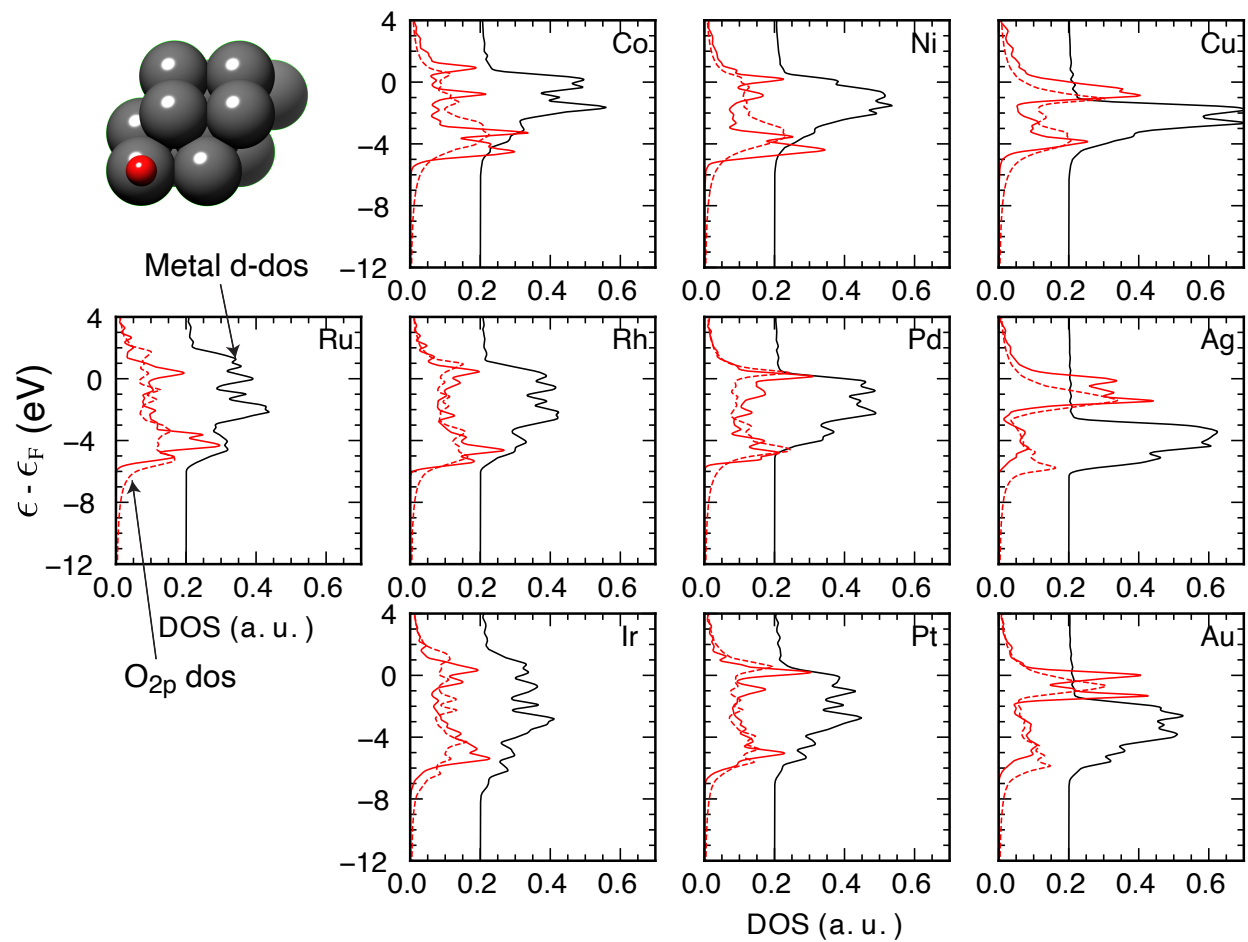
Supplementary Figure 4. Symmetry resolved projected O_{2p} density of states from DFT calculations (solid) and model prediction (dashed) using the Bayesian-learned posterior means of multi-orbital model parameters. Metal d -states are also shown.



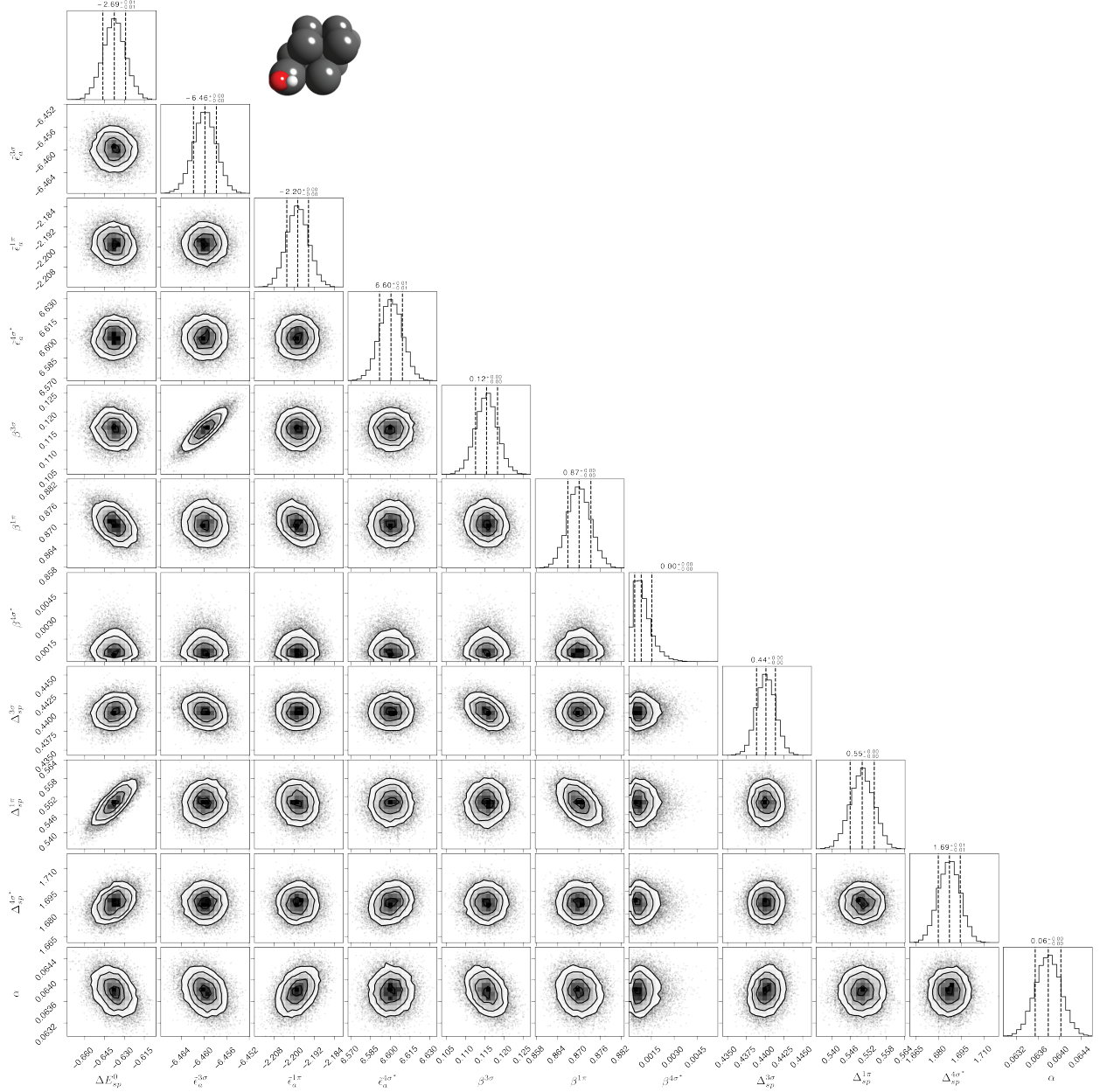
Supplementary Figure 5. The co-variance of the joint posterior distribution for each parameter pair and 1D histogram of single-orbital model parameters for oxygen adsorbed on atop (ΔE_0 , ϵ_a , Δ_0 , α , and β) from MCMC simulations for *O adsorption at the atop site of the close-packed, pristine transition-metal surfaces.

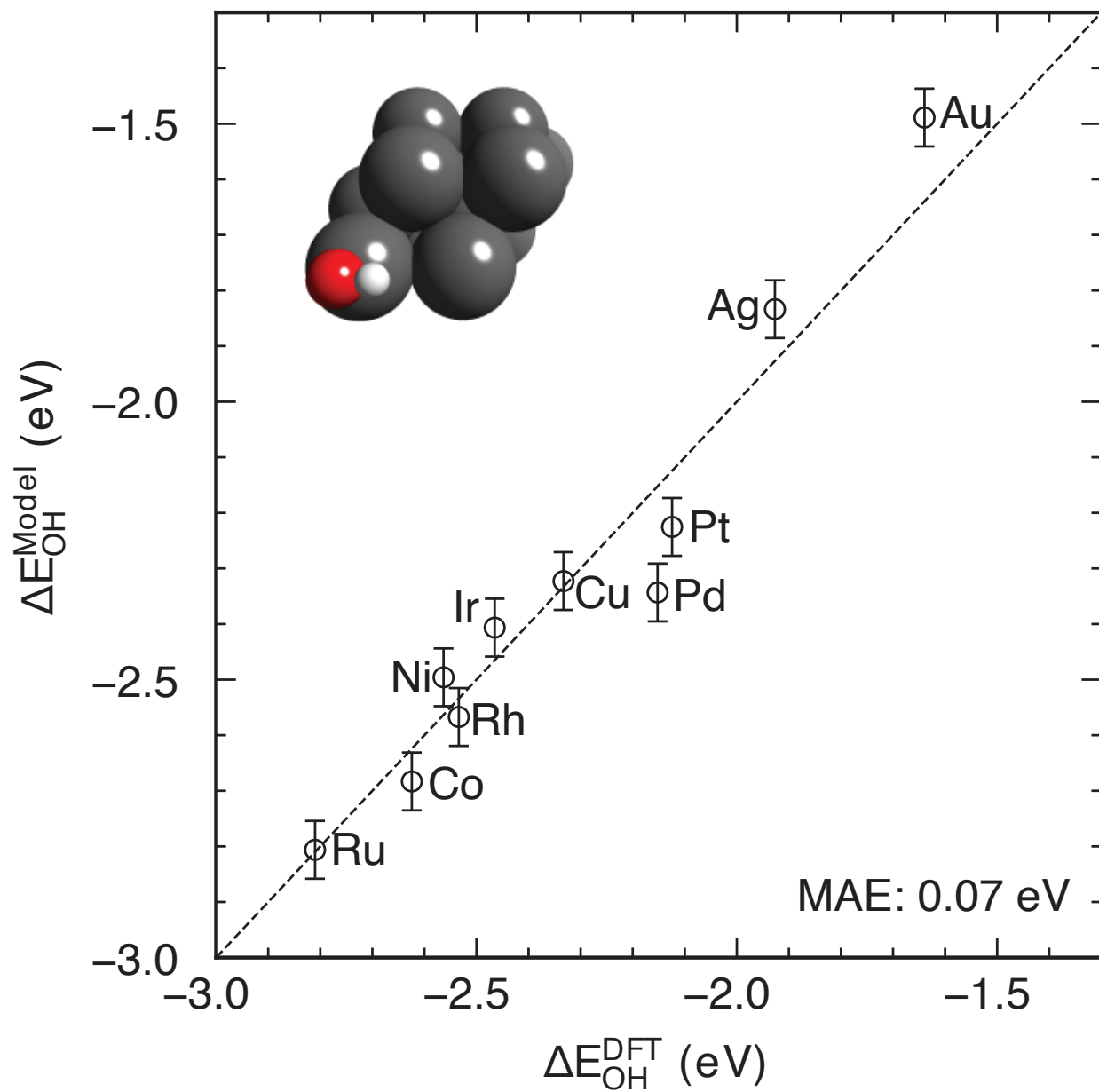


Supplementary Figure 6. DFT-calculated *O adsorption energies at the atop site of metal surfaces vs. model prediction using the posterior distribution of single-orbital model parameters ($\vec{\theta}$, $\vec{\sigma}$). Error bars represent the standard deviation of prediction based on Bayesian sampling.

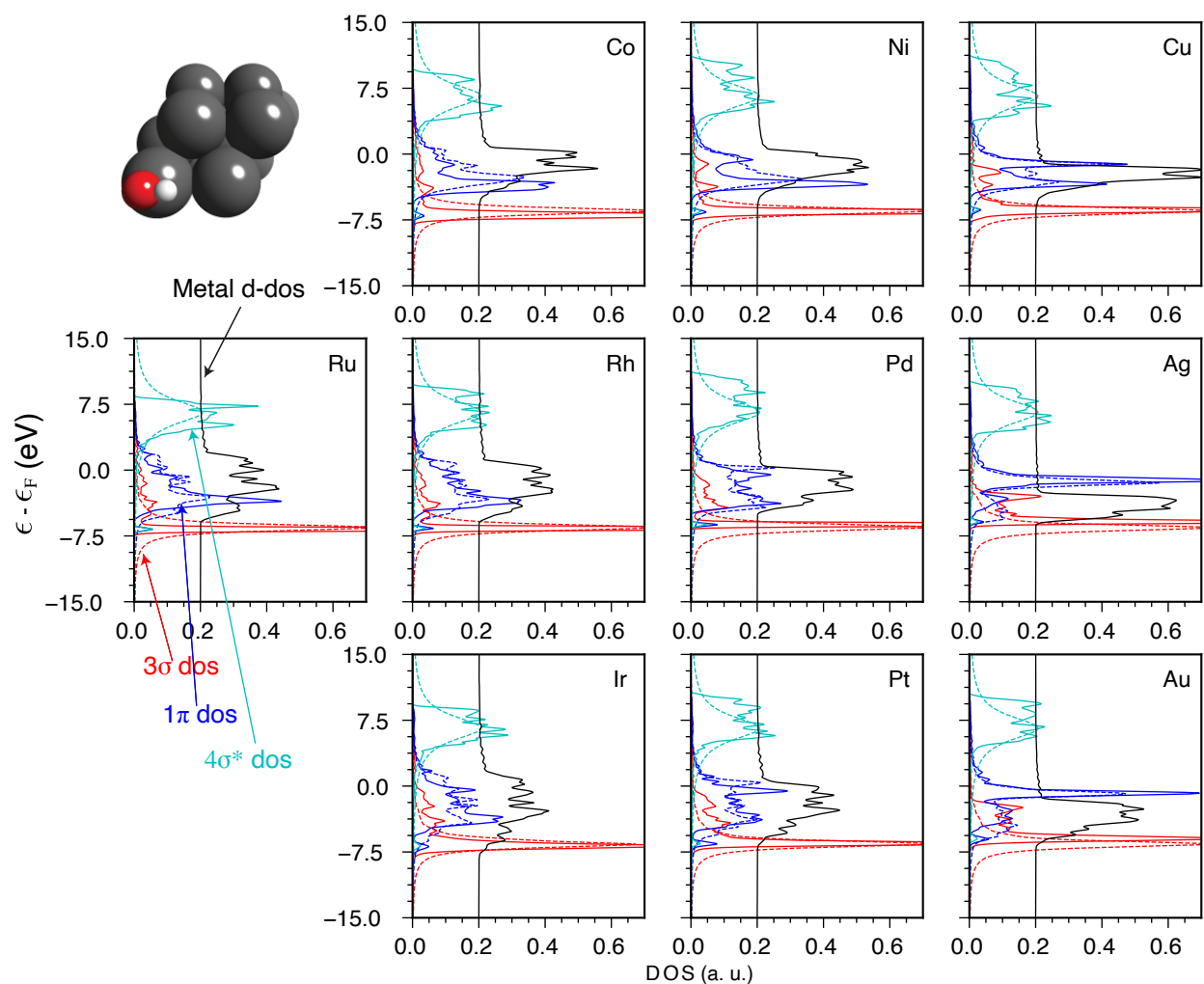


Supplementary Figure 7. Symmetry resolved projected O_{2p} density of states from DFT calculations (solid) and model prediction (dashed) for the atop site using the Bayesian-learned posterior means of single-orbital model parameters. Metal d -states are also shown.

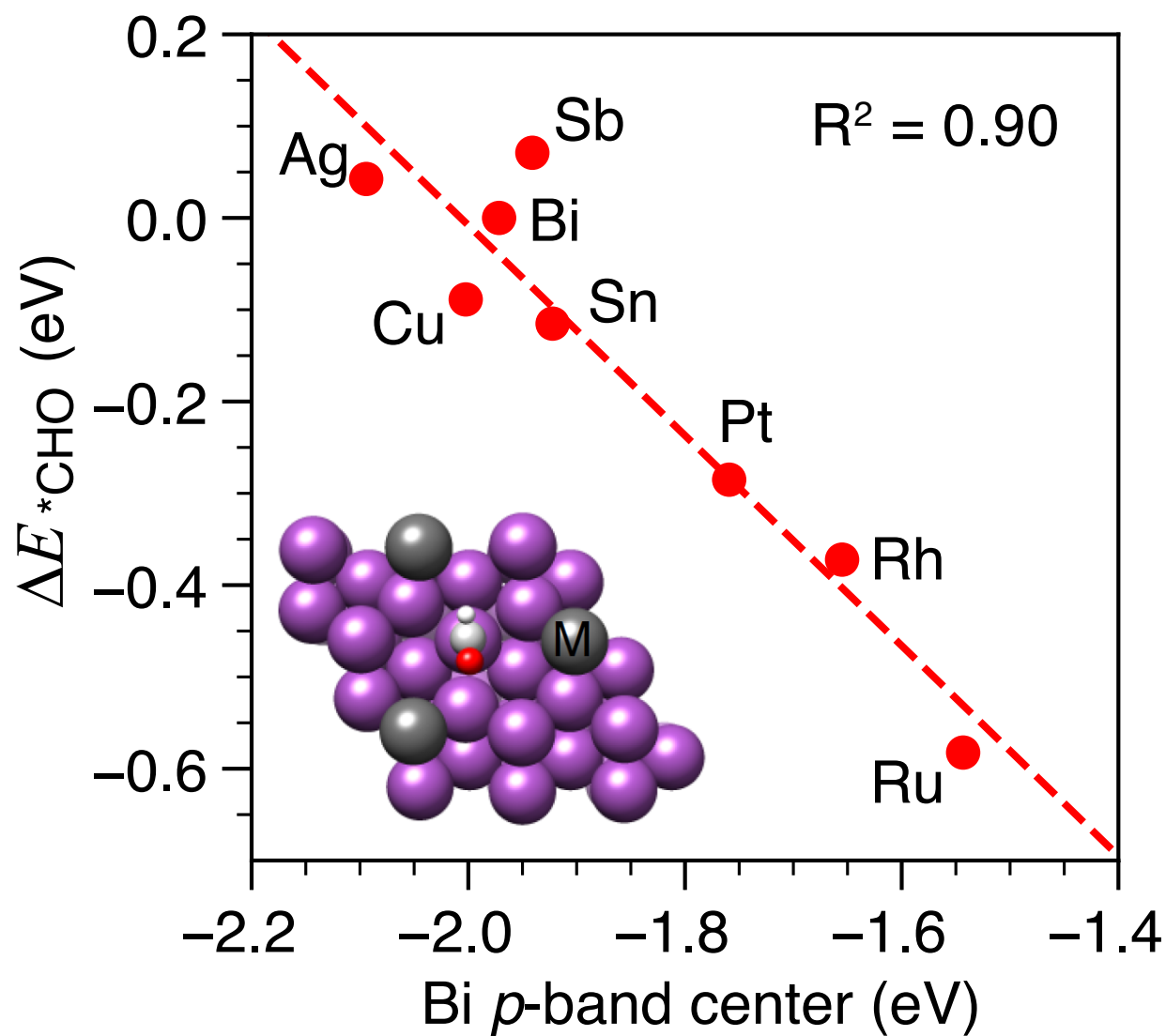




Supplementary Figure 9. DFT-calculated *OH adsorption energies at metal surfaces vs. model prediction using the posterior distribution of model parameters ($\vec{\theta}$, $\vec{\sigma}$). Error bars represent the standard deviation of prediction based on Bayesian sampling.



Supplementary Figure 10. Symmetry resolved projected molecular orbital density of states onto *OH from DFT calculations (solid) and model prediction (dashed) using the Bayesian-learned posterior means of model parameters. Metal *d*-states are also shown.



Supplementary Figure 11. The adsorption energy of $^*\text{CHO}$ at the atop Bi site of M-doped Bi(001) surfaces vs. the Bi p -band center of all states. The inset shows model structures of M-doped Bi(001).

Supplementary References

- [1] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, *J. Phys. Condens. Matter* **21**, 395502 (2009).
- [2] J. P. Perdew, K. Burke, and M. Ernzerhof, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [3] Z. Li, S. Wang, W. S. Chin, L. E. Achenie, and H. Xin, *J. Mater. Chem. A Mater. Energy Sustain.* **5**, 24131 (2017).
- [4] A. Patil, D. Huard, and C. J. Fonnesbeck, *J. Stat. Softw.* **35**, 1 (2010).
- [5] D. Gamerman and H. F. Lopes, *Markov Chain Monte Carlo: Stochastic Simulation for Bayesian Inference, Second Edition (Chapman & Hall/CRC Texts in Statistical Science)*, 2nd ed. (Chapman and Hall/CRC, 2006).