Sette *et al.* Reply: In a Comment on our recent Letter,[1] Chandesris and Rossi (CR) apply a first-neighbor spring-constant model to explain our observation of anisotropic vibrational amplitudes of Cl on Cu(100) and to question our conclusions of comparable anisotropy for the underlying surface-Cu atoms. CR's geometric analysis of effective force constants for various individual atom motions is based on their holding the other atoms in the system stationary. As a consequence, their estimate of the vibrational anisotropy for Cl, $\langle u^2_{\mathrm{Cl}\parallel}\rangle/\langle u^2_{\mathrm{Cl}\perp}\rangle$, implicitly assumes infinitely massive Cu substrate atoms. Given the actual mass ratio of $\frac{35}{63}$, this assumption is questionable at best. Even more serious is that when CR estimate the in-plane nondegeneracy of the surface-Cu displacements, $\langle u^2_{\mathrm{Cu}\parallel 2}\rangle/\langle u^2_{\mathrm{Cu}\parallel 1}\rangle$, they implicitly make the opposite assumption of infinitely massive Cl relative to Cu, and this is clearly wrong. Even if we adopt their first assumption of Cl being light, then the Cl atoms tend to follow adiabatically the Cu vibrations and the proposed in-plane nondegeneracy decreases. This of course weakens CR's objection to our own estimate of the Cl-Cu correlation term. The masses of Cl and Cu are, in fact, close enough so that only a full lattice-dynamical calculation can give accurate predictions of their model for Cl anisotropy and Cu nondegeneracy. Certainly CR's simple and inconsistent estimates cannot.

The implausible consequences of CR's analysis can also be seen by a comparison of their estimates for the vibrational anisotropies of Cl and Cu. For $\langle u^2_{\mathrm{Cl}\parallel}\rangle/\langle u^2_{\mathrm{Cl}\perp}\rangle$ it is $1.67/1.16=1.44$, while for $\langle u^2_{\mathrm{Cu}\parallel 1}\rangle/\langle u^2_{\mathrm{Cu}\perp}\rangle$ it is $1.08/1.6=0.68$. We see that despite the 50% larger force constant assumed by CR for the Cl—Cu vs Cu—Cu bonds, the tightly bound Cl atoms are predicted to move more easily in directions just opposite to those of the more massive Cu. This unphysical result contrasts sharply with the analysis of our experimental data,[1] which shows that the Cl and substrate Cu motions are indeed highly correlated. Therefore, while in our Letter we could not (and did not) make a quantitative assignment for the surface-Cu anisotropy, it clearly must be similar to what was found[1] for Cl, $\langle u_{\mathrm{Cl}\parallel^2}\rangle/\langle u^2_{\mathrm{Cl}\perp}\rangle\simeq 2$.

Aside from CR's inappropriate application of the nearest-neighbor spring-constant model, there is the question of whether the model itself can reliably estimate relative Cl-Cu(100) displacements, i.e., whether it is valid for surface systems. Theoretical and experimental studies[2] of H on Ru(0001), Pt(111), and Rh (001), where the infinitely massive substrate approximation is extremely good, show that this model does not even provide qualitatively meaningful predictions, viz, the actual vibrational anisotropy of H is reversed in all three systems. Therefore, that an approximate treatment of the same model predicts an anisotropy for the Cl-Cu(100) relative motions which happens to be in the same direction as our measured values for the individual Cl-atom displacements can hardly be taken as significant.

Finally, we address the statements made by CR concerning the applicability of a spring-constant model to explain what was reported as surface vibrational anisotropy in an earlier study[3] of Co on Cu(111). Contrary to CR's claim, this system does not simulate an fcc close-packed surface because, while the deposited Co may be epitaxial, it is not commensurate.[4,5] As a result, the distribution of surface Co-Cu force constants leads to correlated motions between Co and Cu that are weaker than between Co and Co. This, coupled with the fact that the individual displacements for Co are known to be smaller than those for Cu, readily explains the reported results and shows that they need not be at all related to surface vibrational anisotropy.[5] The use of a spring-constant model, which presumes the Co-Cu surface force constant to be a simple average of bulk Co and Cu values and which ignores the incommensurability of the adlayer, is therefore inappropriate (and unnecessary) for explaining those results.

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[1] F. Sette, C. T. Chen, J. E. Rowe, and P. H. Citrin, Phys. Rev. Lett. **59**, 311 (1987).

[2] P. J. Feibelman and D. R. Hamann, Surf. Sci. **179**, 153 (1987), and **182**, 411 (1987), and to be published.

[3] P. Roubin, D. Chandesris, G. Rossi, J. Lecante, M. C. Desjonquères, and G. Treglia, Phys. Rev. Lett. **56**, 1272 (1986).

[4] D. Chandesris, P. Roubin, G. Rossi, and J. Lecante, Surf. Sci. **169**, 57 (1986). This work reports Co—Co bond lengths for the adlayer identical to bulk Co, not bulk Cu.

[5] P. H. Citrin, J. Phys. (Paris) Colloq. **47**, C8-437 (1987).