

Dippel *et al.* Reply: We agree with Kaduwela, Van Hove, and Fadley [1] that caution is required in the application of the simple backscattering photoelectron diffraction triangulation method which we have proposed [2] for adsorbate site identification as, indeed, is also required in the interpretation of current holographic reconstructions. In particular, our method will, to some extent, be influenced by scatterers other than the nearest neighbor being probed, *and this possibility was explicitly addressed by us in Ref. [2]*. We dispute, however, that Kaduwela, Van Hove, and Fadley have identified “an important class of realistic bonding geometries” for which such problems do occur.

A realistic assessment of this influence can be obtained either from calculated simulated data for a *possible (performable)* experiment, or from experimental data. We have used both of these methods. Simple single scattering calculations for adsorption on a large cluster of atoms (up to 500) in several geometries [including an fcc(100) surface hollow site] [3], and experimental data for O and CO on Cu(110) [2] all support the contention that our method can identify the nearest-neighbor substrate atoms.

By contrast, Kaduwela, Van Hove, and Fadley [1] show the results of calculations for adsorption on a five-atom (or thirty-atom) substrate cluster for a hypothetical experiment which indicate that the method does not always correctly locate a substrate atom which is either the nearest or second-nearest neighbor. We believe that their calculations do not relate to realistic simulations of likely experiments. First, the cluster size used by these authors may be too small to provide a reliable guide to the true influence of the very many scattering paths which contribute to a real experiment. Truncating a cluster before true convergence can lead to misleading results, although the broad similarity of the thirty-atom substrate results to that of the five-atom cluster suggests that this may not be the dominant problem. More significant, however, is the actual cluster used and the polarization geometry of the model calculation, in which emission is detected along the **A** vector of the radiation and thus at 90° to the incidence direction; this enhances the importance of the second-nearest-neighbor scatterers in the S1-emitter direction by rendering them symmetrically equivalent. Such a geometry optimizes the importance of the effect which Kaduwela, Van Hove, and Fadley seek to demonstrate, but in practice this symmetry is only likely to occur for an adsorbate if the cluster axis is the surface normal; the calculated geometry then precludes measurements in this symmetry direction and so would never be used in a fixed detector geometry experiment (as calculated here and used in our experiments). We further note that the substrate cluster used in Ref. [1] does not correspond to any real surface configuration associated with a simple adsorption structure. In particular, the layer spacing is extremely small, while the first and second neighbors are chosen to have very similar bond lengths. Notice that in

all the examples given of this circumstance in practice [C/Ni(100), N/Ni(100), O/Cu(100), O/Cu(110)] the adsorbate is actually almost coplanar with the top layer, so the actual contribution of the S_2 scatterers in these cases using the polarization geometry calculated in Ref. [3] would be almost zero (identically zero for truly coplanar atoms).

By contrast to these results based on an unrealistic situation, our experimental and theoretical studies using backscattering photoelectron diffraction in its scanned energy mode, applied to more than ten different adsorbate/substrate combinations involving hollow, bridge, and atop adsorption geometries on fcc(100), (110), and (111) surfaces, all show that when photoemission is measured along a nearest-neighbor scatterer-emitter direction, the photoelectron diffraction modulations are dominated by the periodicity associated with the single aligned backscatterer [4]; the group of Shirley at Berkeley has demonstrated this same effect, quite independently, for different adsorption systems [5]. We contend, therefore, that in the face of this extensive *experimental* data base, it seems unreasonable to seriously question our method on the basis of an unrealistic model calculation. We should also note that a recent refinement [6] of our original method, which overcomes the problems of using preselected energies for the polar scan by searching for nearest-neighbor periodicities in coarse energy scans, will greatly reduce the potential problems which Ref. [1] seeks to demonstrate. Finally, we should emphasize that all of the so-called direct methods are intended only to provide a rough guide to the structure in order to reduce the volume of parameter space which needs to be searched in structural refinement by existing modeling techniques.

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