Comment on "In-Plane Lattice Reconstruction of Cu(100)"

A recent structural analysis using low-energy electron diffraction (LEED) by Müller et al. [1] reports that the outermost layer of clean Cu(001) has an in-plane lattice parameter contracted relative to the bulk lattice by 1% due to tensile stress. This result is surprising as the structure of this surface has been well studied experimentally and theoretically, without previous evidence of in-plane reconstruction ([2,3], and references therein). Furthermore, an in-plane contraction is in conflict with our results from high-resolution LEED (HRLEED), x-ray diffraction (XRD), and scanning tunneling microscopy (STM).

Müller et al. [1] recognize that a contraction within the outermost layer requires defects in the long-range order to accommodate the surface to the bulk lattice. They suggest a length scale of 250 Å, which is greater than the transfer width of their LEED optics. Our HRLEED results, however, reveal that there are millimeter size regions on Cu(001) at $T = 213$ K where the in-phrase width of specular diffraction is resolution limited, implying atomic order averaging over 1000 Å [4]. A layer with in-plane contraction will no doubt lose atomic order at a step; however, even surfaces with controlled step separations ranging from 30 to 700 Å retain resolution limited atomic order.

Long-range order in a surface layer with a lattice contraction implies that a large fraction of the surface atoms will not be aligned with the substrate. This is not supported by total yields observed with ion scattering at either 107 or 305 K [2,3]. Misalignment of surface atoms should also be reflected in vertical heights, since atomic registry for a uniformly contracted surface, as proposed by Müller et al. [1] will range from fourfold hollow to on-top positions. Vertical heights would be expected to vary about 0.75 Å (using a hard-sphere model) and form a square moiré pattern across the surface. Instead, our STM images at 213 and 300 K reveal flat surfaces with less than 0.1 Å corrugation across terraces as large as 2000 Å [5].

An in-plane contraction should also produce additional diffraction peaks. While not directly observable with conventional LEED, a 1% variation in lattice parameter is easily resolved using XRD. Since x rays are sensitive to both surface and near-surface layers, the model of Müller et al. [1] should lead to two diffraction peaks. In a recent XRD study of Cu(001) using synchrotron radiation [6], a number of surface sensitive diffraction rods were measured. Only one peak, which is well fit by a single Lorentzian, was observed in both transverse and radial scans within ±1% of the bulk truncation rod. Although a poorly ordered surface may contribute only as a broad background in XRD, the HRLEED data have eliminated this possibility. The reciprocal space positions of the observed diffraction peaks are very near integer values, i.e., nearly aligned in plane with bulk Bragg diffraction. Specifically, a Lorentzian fit to the rod near $(1, 1, 0.1)$ indicates a surface in-plane lattice parameter within 0.07% of the bulk. Uncertainty originating from error in crystal alignment should be less than 0.1%.

The array of experimental evidence supporting a commensurate surface layer with bulk in-plane spacing suggests that further examination of the LEED experiments and analysis of Müller et al. [1] be made for possible systematic error. For example, the best-fit bulk lattice spacing normal to the surface (1.79 Å) produced by the LEED analysis of Müller et al. also shows approximately a 1% contraction [7]. This homogenous contraction supports concern that a systematic error is responsible for the apparent in-plane contraction. Furthermore, recent structural studies of Cu(001) have shown an unusually large in-plane root-mean-squared (rms) motion of surface atoms [2,3,6]. It has been previously reported that rms motion can affect LEED intensities in a way which mimics a static in-plane displacement of the outermost layer at a surface [8].

A. P. Baddorf, A. K. Swan, and J. F. Wendelken
  Solid State Division, Oak Ridge National Laboratory
  Oak Ridge, Tennessee 37831-6030

[1] S. Müller, A. Kinne, M. Kottcke, R. Metzler, P. Bayer, L. Hammer, and K. Heinz, Phys. Rev. Lett. **75**, 2859 (1995).

[2] Q. T. Jiang, P. Fenter, and T. Gustafsson, Phys. Rev. B **44**, 5773 (1991).

[3] D. E. Fowler and J. V. Barth, Phys. Rev. B **52**, 2117 (1995).

[4] J.-K. Zuo and J. F. Wendelken, Phys. Rev. Lett. **70**, 1662 (1993).

[5] A. K. Swan, Z.-P. Shi, J. F. Wendelken, and Z. Zhang (to be published).

[6] A. P. Baddorf (to be published).

[7] Thanks to Shobhana Narasimhan for pointing this out to us.

[8] A. F. Wright and D. C. Chrzan, Phys. Rev. Lett. **70**, 1964 (1993).