Direct Observation of Atomic Steps in the Surface Reconstruction of the Pt (110) Plane

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Atomic steps involved in the reconstruction of the Pt (110) plane, from the (1×1) to the (1×2) structure, have been directly observed in the field-ion microscope with use of nanosecond-pulsed-laser heating of the surface. The dominant steps are breaking of [110] atomic rows into fragments of two to a few atoms and lateral and cross-channel jumps of these fragments. Within the ~ 5-ns time resolution of the experiment, atoms are seen to jump in pairs or groups, and jumps of single atoms are rarely seen. The (1×2) surface is stabilized by an attractive adatom interaction at ~ 2.8 and ~ 8 Å, and a repulsive interaction at intermediate distances.

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Recently heating of solids by pulsed laser beams of very short duration has attracted much attention.[1] This method provides a unique opportunity to study the thermodynamics of transient phase transitions and kinetics of regrowth and phase transformation on extremely short time scales and in extreme conditions. It is now possible to study time dependence of the creation of electron-hole pairs in solids (electron heating), the subsequent transfer of energy into the lattice, and melting and vaporization, etc., with subpicosecond time resolution.[2,3] We report here a preliminary attempt to observe directly in the field-ion microscope the detailed atomic steps involved in surface reconstructions and atomic structures of transition states using nanosecond-pulsed-laser heating. One advantage of pulsed-laser heating as compared to the conventional methods of heating in a study of surface atomic processes is that the surface can be quenched with a rate greater than 10^{11} K/s after the heating.[1-4] Thus some of the transition states which are difficult to observe by ordinary methods may be frozen for microscopic observation.

In this first study we focus on the (1×1) to (1×2) surface reconstruction of the Pt (110) plane for the reason that this reconstruction is now fairly well understood.[5] LEED, atomic-beam scattering, transmission electron microscopy, and scanning tunneling microscopy studies of fcc (110) surfaces support a missing-row model.[5,6] This model has also been studied by a direct field-ion microscope (FIM) observation.[6] In that FIM study using a conventional resistivity heating technique, a (1×1) surface of Pt (110) produced by 78-K field evaporation is found to reconstruct to a (1×2) structure above 310 K. The heating method has a quenching rate of ~ 10 K/s and thus no detailed atomic steps of the reconstruction or atomic structures of the transition states can be observed or have been reported. This study of ours is intended to reveal these atomic steps and atomic structures, and also to demonstrate the usefulness of the pulsed-laser tech-

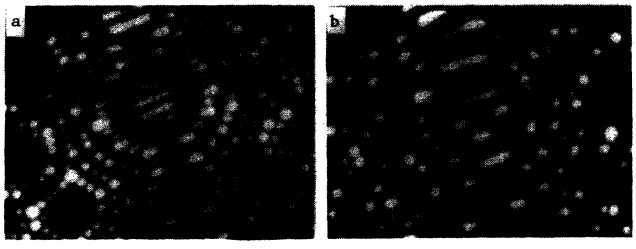

FIG. 1. He FIM images of a Pt (110) plane. (a) (1×1) structure produced by low-temperature field evaporation. (b) (1×2) structure produced by pulsed-laser heating of the same surface. All FIM images are taken at 20–35 K.

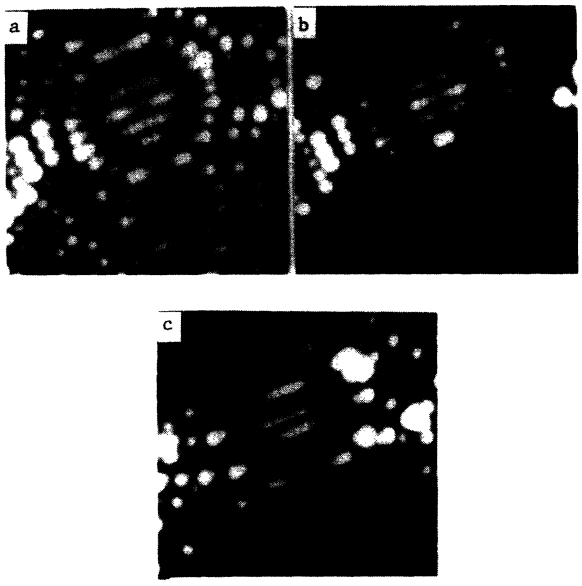

FIG. 2. Within a resolution of ~ 5 ns, a row of several atoms are seen to jump together by irradiation of *one* laser pulse. Temperature is about 500 K.

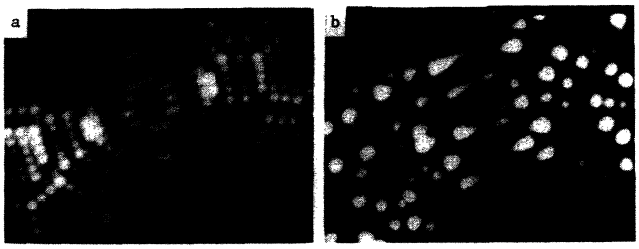

FIG. 3. (a) A (1×1) surface. (b) Structure of a transition state induced by pulsed-laser heating.

nique in the study of atomic structures of transition states in phase transitions.[1]

Procedures and vacuum requirements in FIM study of surface atomic processes are well established and can be found in the literature.[7] A tip is first field evaporated at ~30 K. Helium field-ion images show an atomically well-developed surface of the emitter which exhibits a (1×1) structure at the (110) plane. The tip and channel plate voltages are then turned off and one to a few laser pulses are then fired to heat the surface. During the rapid heating and cooling of the surface there is no applied field. Voltages are then carefully raised again to reveal the atomic structures of the surface. As our system is not yet automated, this procedure takes 5 to 15 s. The laser pulses, 5 ns in width and 2 mJ/pulse, come from a nitrogen laser of $\lambda = 337$ nm and are focused to the very end of the tip with a lens mounted on an *XYZ* translator.[8] The final temperature reached by the surface is estimated from the reduction of the field evaporation voltage to be ~400 to 600 K. For this study the exact final temperature reached by the surface is not essential since there are still many unknowns in what effect really produces the atomic motions in our observation. It is possible that electron heating and direct quantum excitations are involved in these atom jumps.

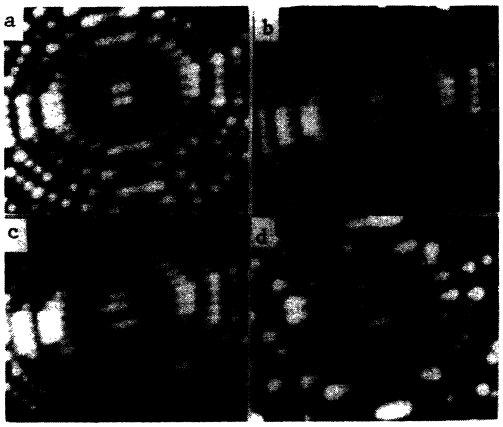

FIG. 5. Another example similar to that of Fig. 4.

In Fig. 1(a) a small (1×1) Pt (110) plane of 18 atoms, produced by 30-K field evaporation, is shown. The structure of the underlying layer is also (1×1). The unit cell has a rectangular structure of 2.77×3.92 Å. After the surface is irradiated with laser pulses, a much more stable surface with respect to surface heating as shown in Fig. 1(b) is formed which has the (1×2) structure for both the top layer, which still has 18 atoms, and the edges of the second and third layers. Field evaporation reveals that for the second and third layers, surface reconstruction occurs only at the edge,

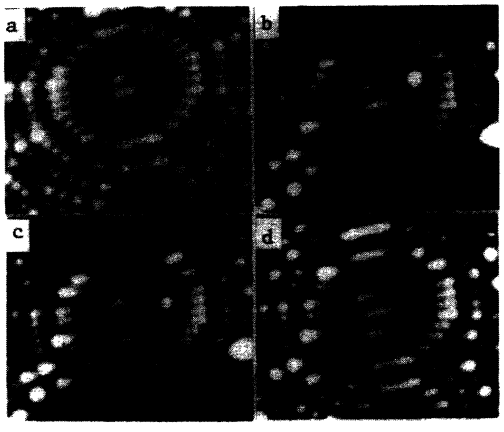

FIG. 4. Detailed steps in the (1×1) to (1×2) surface reconstruction. Between two images is the irradiation of *one* laser pulse of 5-ns width.

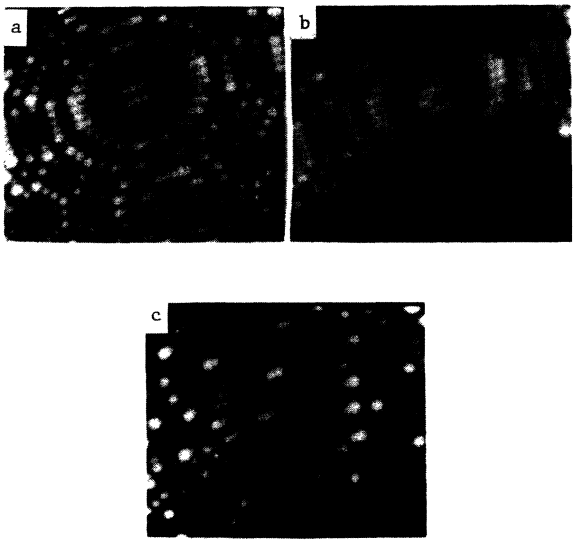

FIG. 6. Another example similar to that of Fig. 4.

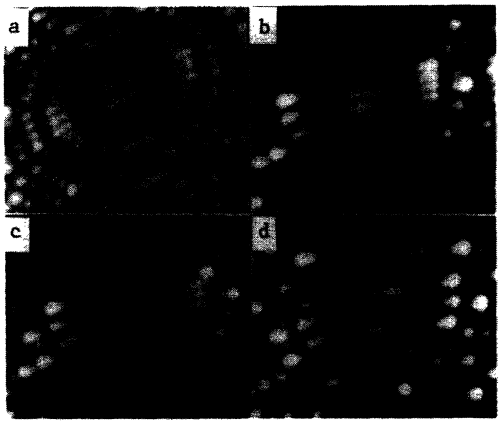

FIG. 7. Another example similar to that of Fig. 4.

of two to four atoms width. The underlying layers are still (1×1); thus our result is consistent with the "simple" missing-row model, but not with other models which should show two to three reconstructed layers. Atom transport is seen to occur in nearest-neighbor pairs or in small [110] atom rows of several atoms as is clearly seen in Figs. 2(a) and 2(b). In Fig. 3(b) atomic structure of a transition state is shown. Closely packed [110] atomic rows are broken up in small fragments by both lateral motion in the [110] direction and cross-channel motion in the [001] direction of pairs or rows of atoms. Each atom-row fragment consists of mostly two and sometimes more atoms. Single atoms are rarely found. Before a perfect (1×2) surface is formed, the plane may be intermixed with atomic rows separated by single [110] spacing, or double spacing as shown in Fig. 2(c). In these experiments, if the laser power is high then one or two extra atoms will occasionally appear on the top surface layer. These atoms are activated from the interior of the second layer. Occasionally we may lose one or two atoms from the top layer by their falling off the lattice step, by accidental field evaporation (which is unavoidable in order to bring out a good field-ion image), and in rare cases by recombination of an adatom-vacancy pair as shown in Figs. 4(b) and 4(c).

To study in better detail the atomic steps in the surface reconstruction, we have observed the behavior of

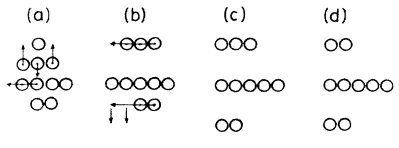

FIG. 8. Suggested atomic steps for the reconstruction shown in Fig. 4.

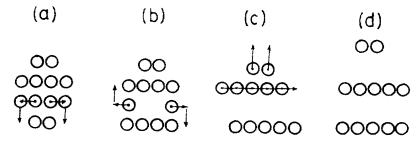

FIG. 9. Suggested atomic steps for Fig. 5.

small planes of ~4 atomic rows and 10 to 15 atom sizes in the reconstruction process. Figs. 4–7 give four examples of these reconstructions. Based on observation of these and many others, general rules of atom transport and detailed atomic steps of surface reconstruction can be deduced, of course within the ~5-ns time resolution of the experiment. Figures 8–11 are suggested to be responsible for the reconstructions shown in Figs. 4–7. Atomic movements often occur suddenly, and even within 5 ns many changes can be seen. It is still possible that these observed steps involve sequential jumps of single atoms, especially in the cross-channel jump of small atomic rows. We conclude that a (1×1) macroscopic-size Pt (110) plane consists of small patches of well-ordered (1×1) islands. The $(1\times1)\rightarrow(1\times2)$ transition involves breaking of [110] atomic rows and lateral and cross-channel jumps of these fragmented atomic rows of two to several atoms.

In summary, using nanosecond-pulsed-laser heating we are able to observe atomic structures of a transition state and also atomic steps in a phase transition for the first time. In the (1×1) to (1×2) surface reconstruction of the Pt (110) plane, the (1×2) structure is stabilized by an attractive interaction of the adatoms at ~2.8 Å and at ~8 Å, and also by a repulsive interaction in the intermediate distances, similar to what Casanova and Tsong have measured for adatom-adatom interactions on the W (110) plane.[9] Thus this study gives further evidence of the nonmonotonic distance dependence of adatom-adatom interactions.[10] It is also interesting to note that within the time resolution of the present study, ~5 ns, Pt atoms on the Pt (110) are seen to jump together, in great contrast to the cooperative walk of atoms in the neighbor channels of the W (112) plane where atom chains always move by jumps of individual atoms.[11,12] The com-

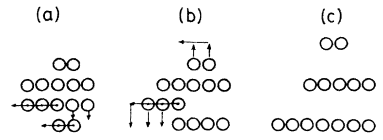

FIG. 10. Suggested atomic steps for Fig. 6.

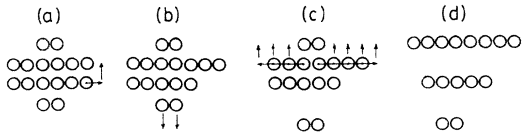

FIG. 11. Suggested atomic steps for Fig. 7.

bination of a pulsed-laser technique and high-resolution microscopy should be able to reveal the atomic steps involved in structural phase transition, kinetics of atom transport, and the atomic structures of the transition states.

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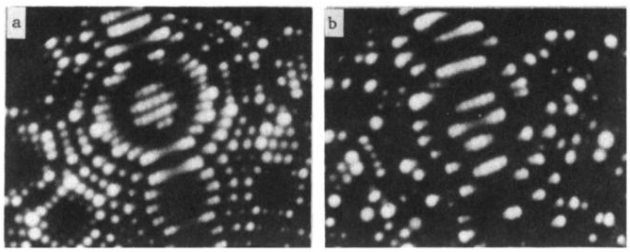

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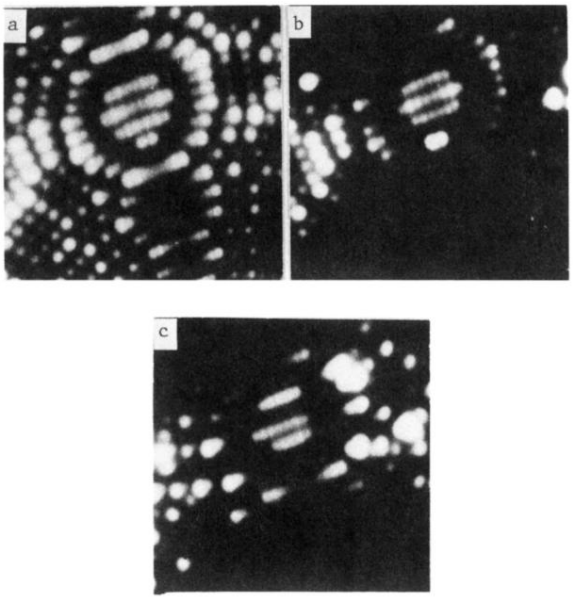

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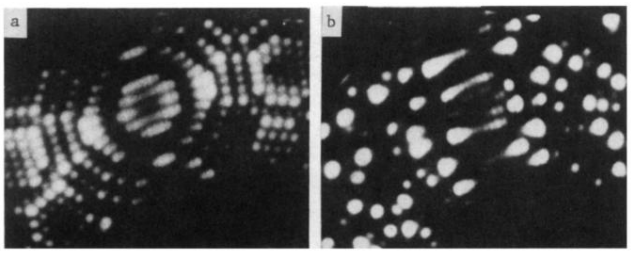

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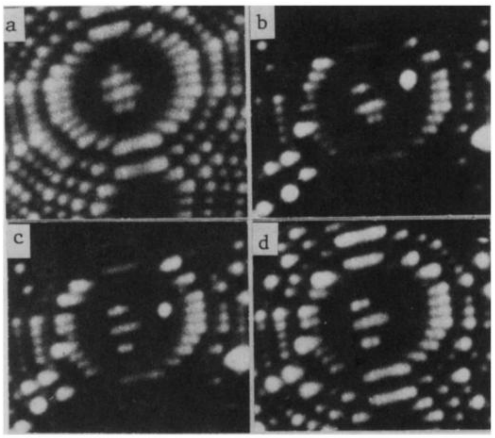

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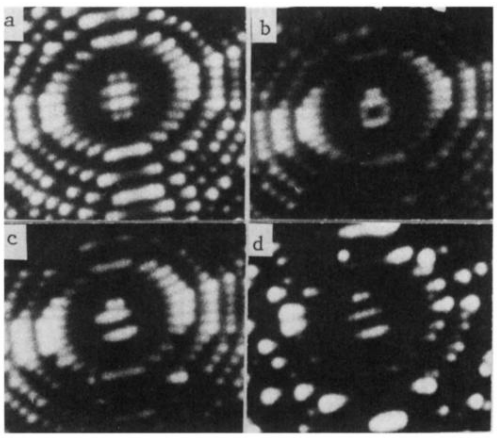

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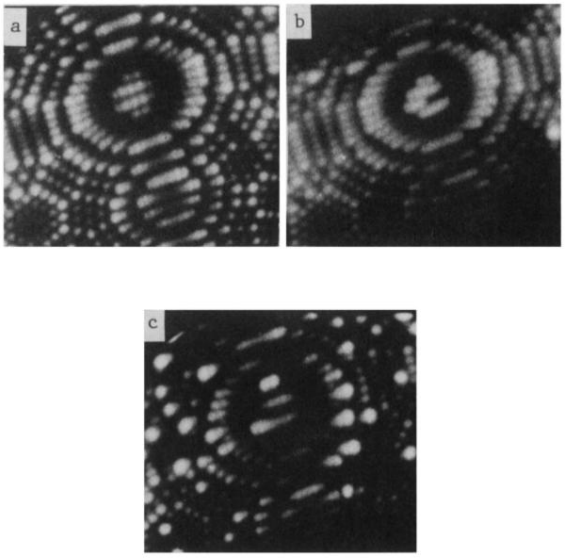

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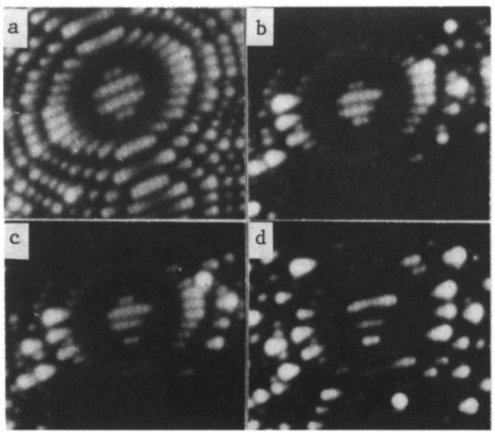

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