Cross-sectional time-resolved high-resolution transmission electron microscopy of atomic-scale contact and noncontact-type scannings on gold surfaces

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Atomic-scale contact and noncontact scannings on gold surfaces were performed by gold tips in time-resolved high-resolution transmission electron microscopy (HRTEM) using a piezodriven specimen holder. The structural variations of atomic arrangements in the scanning process were observed *in situ* with a time resolution of 1/60 s. Possibility of combination-type microscopy of HRTEM and scanning-probe microscopy was shown. Mechanical processing at one atomic-layer scale was demonstrated. [S0163-1829(97)50412-7]

The elucidation of mechanical interaction between two materials at atomic scale has recently been an important subject for such studies of scanning probe microscopy (SPM) as scanning tunneling microscopy (STM) and atomic force microscopy (AFM). In AFM, images of surface structures of the specimens are reconstructed from the atomic interaction between surfaces of needles and specimens.[1] The interaction is affected by the shapes of needles and surfaces, as well as the distance, stress, or chemical reaction between the needles and the surfaces. In particular, in contact-type AFM, problems of the atomic interaction are rather serious because the structures of the surfaces are modulated by the scanning.[2] The interaction at the scanning should be elucidated in order to understand properly SPM images in relation with the factors.

In the present study, atomic-scale contact and noncontact surface scannings were performed on gold surfaces by gold tips in time-resolved high-resolution transmission electron microscopy (HRTEM) using a piezodriven specimen holder. The structural variations of atomic arrangements in the scanning process were observed *in situ* at atomic scale and at time resolution of 1/60 s.

Figure 1 shows an illustration of a specimen holder of HRTEM for the present nanometer scale surface scanning. The mobile side is connected with a tube-type piezoelectric device for fine displacement and a microscrew motor for coarse displacement. The specimen for needle at the mobile side is mounted on the tip of a lever connected with the piezodevice. The mobile side displaces along the x direction up to ± 1 mm by the motor. The fine displacement along the x direction is adjusted by homogeneous elongation and shrinkage of the piezodevice. The fine displacements along the y and z directions are caused by elongation and shrinkage at one side of the tube-type piezodevice. The resolution of three-dimensional displacement by the piezodevice is described later.

The specimens used in this work were polycrystalline gold wires of 0.1 mm diameter and 5 mm length. The tips of the wires were thinned by irradiation of Ar^+ ions accelerated at 3 kV. The two needle-shaped specimens were mounted separately at the fixed and mobile sides of the holder. The tips of the specimens in the mobile side and the fixed side were used for a scanning needle and a surface, respectively. The specimen holder was installed in a 200-keV high-

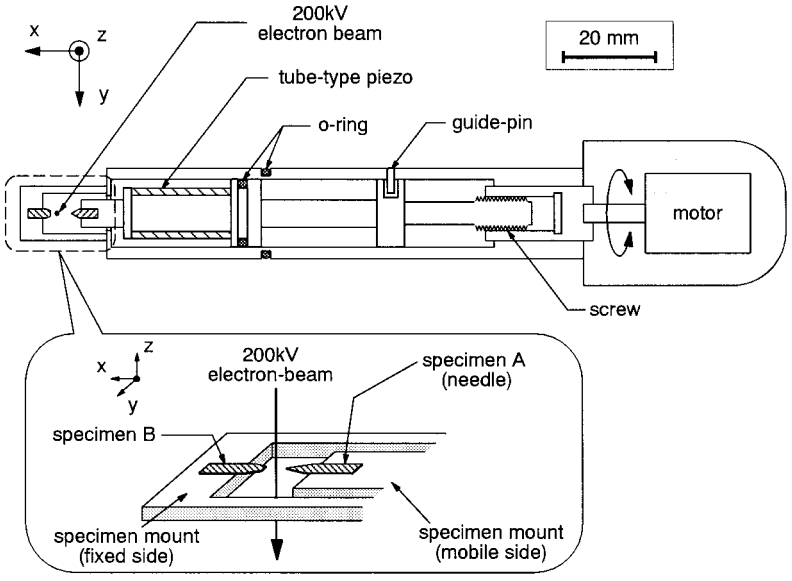

FIG. 1. Illustration of a specimen holder of a high-resolution transmission electron microscope for the present atomic-scale surface scanning.

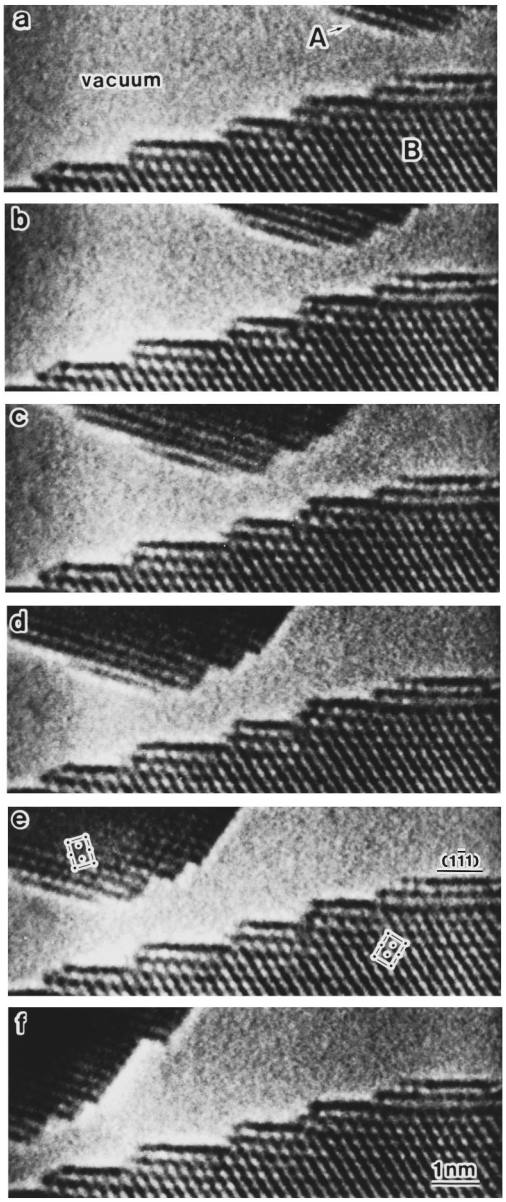

FIG. 2. Cross-sectional time-resolved high-resolution images of scanning of a gold tip in the mobile side (*A*) along a surface of a gold tip in the fixed side (*B*). The distance between the two tips is kept at 1.0 nm. Frames show the unit cells of gold with face-centered-cubic structure projected along the [110] direction. Time is (a) O s, (b) 1 s, (c) 2 s, (d) 4.5 s, (e) 6 s, and (f) 8 s.

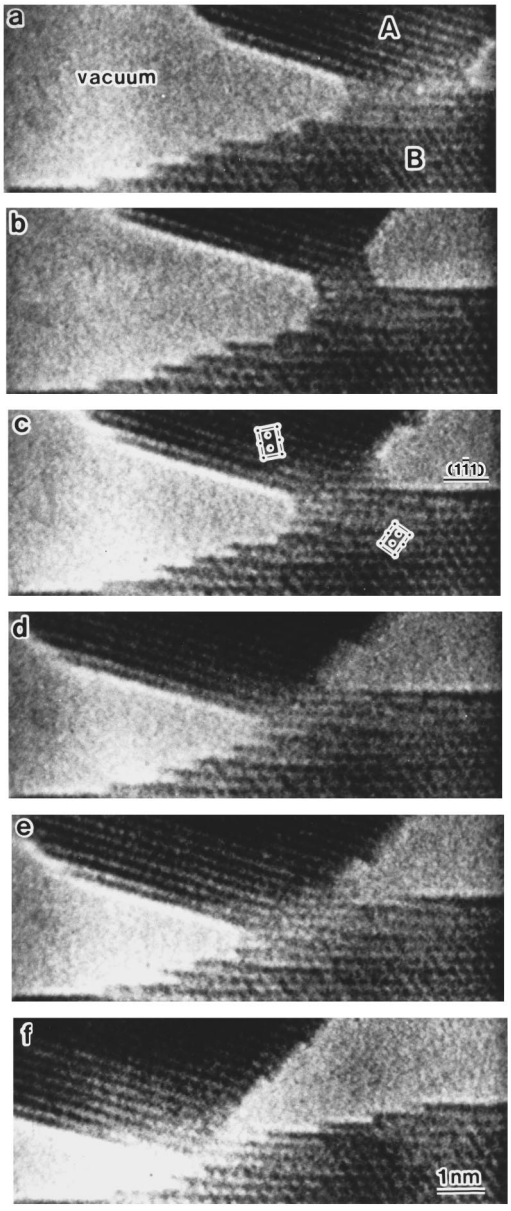

FIG. 3. Cross-sectional time-resolved high-resolution images of the scanning of a gold tip of the mobile side (*A*) when the distance between the tip and a gold tip in the fixed side (*B*) is 0 nm. The two tips bond by a boundary of a few atomic columns width. Frames show the unit cells of gold with face-centered-cubic structure projected along the [110] direction. Time is (a) O s, (b) 3 s, (c) 8 s, (d) 11 s, (e) 16 s, and (f) 19 s.

resolution transmission electron microscope (JEOL, JEM-2010). Surface scanning was carried out in the microscope at ambient temperature. Structural variations of atomic arrangements through the scanning processes were observed *in situ* using a SIT-TV camera and recorded with a digital video tape recorder at the time resolution of one field of video images, 1/60 s.[3,4] Lattice resolution and point-to-point resolution in real space using the present specimen holder was 0.14 and 0.20 nm, respectively. Electron beam irradiation density at the observations was 31×10^4 A/m^2.

Maximum displacements along the *x* and *y* directions were 1.2 and 11.4 μm, respectively, when 150 V was applied to the piezodevice. The mobile side can be displaced up to ±1 mm along the *x* direction by the microscrew motor. Maximum displacement along the *z* direction was estimated to be 11 μm judging from the variation of the focusing current of an objective lens of the microscope at just focus condition. Differences of positions due to the hysteresis along the *x* and *y* directions were 1.3 and 2.5 nm, respectively, when the tip was displaced by 100 and -100 nm.

The minimum steps of the displacement along the *x* and *y* directions were 0.16 and 0.22 nm, respectively. The steps are similar to the spacings of {220} and {111} lattice planes of gold with a lattice constant of 0.408 nm. Applied voltages at one step along the *x* and *y* directions are 28 and 3.5 mV, respectively. The minimum step along the *z* direction was estimated to be similar to that along the *y* direction, because

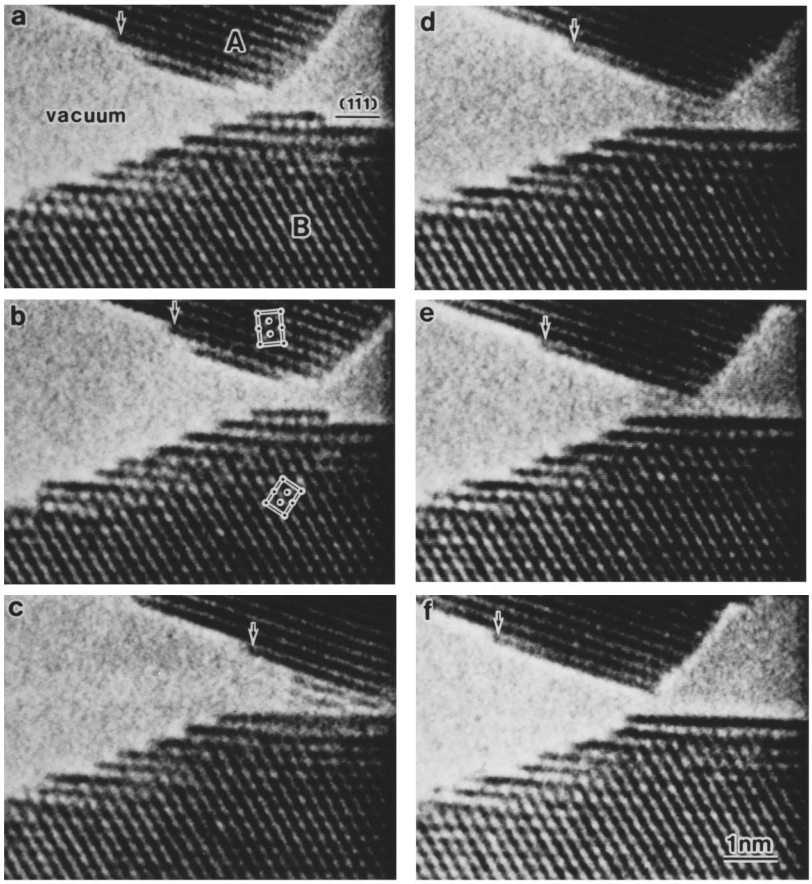

FIG. 4. Cross-sectional time-resolved high-resolution images of displacement of a gold tip in the mobile side (*A*) on a surface of six atomic columns width in the fixed side (*B*). Frames show the unit cells of gold with face-centered-cubic structure projected along the [110] direction. Each arrow in the images shows the same position in the tip on the mobile side.

the mechanism of the displacements along the *y* and *z* directions is the same. The steps along the three directions are enough to scan surfaces of gold at atomic scale.

The relative height of two tips is adjusted before the surface scanning. High-resolution images of edges of the tips vary with the thickness of the tips and the defocus condition. The thickness is usually almost the same after the ion milling. Similar high-resolution images emerge at the edges at various defocus conditions when the relative height is the same. Thus the variation of the relative height can be confirmed by the displacement of the tip in the mobile side along the *z* direction and the subsequent high-resolution observation on a television monitor. It was found from our previous experiments that the relative height can be reduced to less than 8 nm. Finer estimation of the relative height will be performed by a comparison of through-focus series of actual and simulated high-resolution images of surfaces using the multislice method.[5]

Some tips appeared on an uneven edge of each specimen after the Ar^+ milling. The tips were oriented randomly because the specimen used was polycrystalline gold. We can thus find the tips whose directions are suitable for high-resolution observation, or adjust the direction easily by tilting the specimen at the *x* direction. The [110] axes were selected to be parallel to the incident electron beam direction in this work.

Figure 2 shows a series of cross-sectional time-resolved high-resolution images of scanning of a gold tip in the mobile side (*A*) along a gold surface of a tip in the fixed side (*B*). The distance between the two surfaces is kept at 1.0 nm. No contamination is observed in the surfaces. It is demonstrated that the tip in the mobile side can be displaced to the left along atomic steps of the top surface of the tip in the fixed side. The surface structure is not varied by the displacement. The operation of the tip is similar to the surface scanning of a needle in noncontact-type AFM.

Figure 3 shows a series of time-resolved high-resolution images of the scanning of a tip of the mobile side when the distance between two tips is 0 nm. The two tips bond by a grain boundary of a few atomic columns width. Stick-slip motion is observed at the displacement. This means that the displacement is disturbed by a kinetic friction. The formation of the grain boundary and stick-slip motion were not observed when the two tips do not make contact but separate along the *z* direction. The surface structure varies by the displacement. The displacement corresponds to the fracture phenomenon in surface scanning in contact-type AFM.

It is clear from the observation that several layers at the two surfaces and the contact boundary are responsible for the contact-type surface-scanning process. Therefore, the strength of the boundary is determined by the condition near the top surfaces. The strength of the boundary is attributed to a static friction when the two tips bond and fix. The strength of the boundary is responsible for a kinetic friction during the displacement.

Figure 4 shows a series of time-resolved high-resolution images of the displacement of a tip of the mobile side on a surface of six atomic columns width in the fixed side. No structural variation is observed when the two tips do not make contact [Figs. 4(a) and 4(b)]. The top surface of six atomic columns width in the fixed side [Figs. 4(a) and 4(b); *B*] is modulated and the neck-shaped grain boundary is

formed when the tips make contact [Fig. 4(c)]. This shows that the neck-shaped boundary is formed through atomic diffusion at the surface. The two tips connect by the neck-shaped boundary of four atomic columns width at the displacement [Figs. 4(d) and 4(e)]. The top surface of six atomic columns width appearing in Figs. 4(a) and 4(b) disappears after the displacement [Fig. 4(f)]. The observation shows that a kind of mechanical processing of one atomic layer can be performed using the present method.

No contamination is observed in the surfaces as shown in Figs. 2–4, although gaseous molecules, such as oxygen and nitrogen, might absorb before the observation. The present specimen, gold, has low activity and the thickness of the contamination is inferred to be a few atomic layers or less. Such molecules would desorb due to electron beam irradiation. If other active materials are used as specimens, contamination and/or reaction layers that disturb surface structural analyses will be observed on surfaces. Specimen preparation and transfer chambers attached with electron microscopes will be useful for the observations of the surface-scanning process of such active materials.[6]

Height control along the *z* direction is important for studies of interaction of the tips, such as attractive, unstable, and repulsive regions of potential. The relative height can be reduced less than 8 nm before the contact of the tips, as already described. We can also confirm the contact of the tips by the direct observation of the formation of grain boundary and stick-slip motion. The relative height is assumed to be zero at separation after the contact. Consequently, the height can be adjusted at the resolution of the displacement along the *z* direction after the contact.

The present observation also shows the possibility of real-space atomic observation of surface structures by the combined use of HRTEM and STM. The merits of the combined use are as follows. It is possible to (1) select clean and flat surfaces for STM, (2) evaluate the shape of a needle for STM, (3) monitor *in situ* the position and movement of the needle, (4) analyze the internal structure near the surface, and (5) apply various kinds of analytical methods relating to HRTEM, such as EDX or EELS from nanometer area at surfaces. It is expected that the method gives information which cannot be obtained from only HRTEM or only STM.

Similar combination-type microscopes of reflection electron microscopy (REM) and STM or TEM and STM have been attempted by Kuwabara *et al.*,[7] Spence *et al.*,[8–10] and Lutwyche *et al.*[11] However, surface scanning by the tips still has not been imaged successfully at atomic resolution by the REM or the TEM. In the present study, the surface scanning is clearly observed at atomic scale. Surface structure analysis by STM will indeed be performed using the present specimen holder, if the piezodevice is driven by controlling the tunneling current between the two tips at current-constant mode. The present observation thus shows an interesting possibility to combine HRTEM and STM.

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