Highly sensitive reflection high-energy electron diffraction measurement by use of micro-channel imaging plate

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Micro-channel plate (MCP) was used to intensify the image on the fluorescent screen in reflection high-energy electron diffraction. The primary electron current could be reduced by four orders of magnitude to get the image of the same intensity as in the measurement without MCP. The reduction of electron beam enabled observation of ionic crystal surfaces without causing electrolysis and charging effect. © *2000 American Institute of Physics.* [S0034-6748(00)02309-1]

I. INTRODUCTION

The epitaxial thin film growth is of great importance from viewpoints of basic research and technical applications. New functional materials could be obtained in the form of ultrathin films with novel phases in crystal structure, crystal face, etc. Although intensive works have been carried out so far for preparing semiconductor and metal films, fewer works have been done on ionic compounds.[1,2] Recent development in the fields of oxide superconductors and wide band gap semiconductors, however, require the knowledge on the preparation method for materials, in which ionic bonds play an important role.

Reflection high-energy electron diffraction (RHEED) is a popularly used method to determine morphology and crystallographic orientation of film surfaces during growth. In addition to the static image, the intensity oscillation of diffraction spots provides information on the growth process. In the case of ionic crystals, however, there are some problems in the RHEED analysis. One is the problem of charge buildup, because most of ionic crystals are electrically insulating. In many cases it is impossible to continue the RHEED observation, because charge buildup on the specimen surface repels the probing electrons. In case that the RHEED observation is possible due to ionic conductivity or surface conductivity, there appears another problem related with electrolysis on the ionic crystal surface. For example, prolonged electron impingement causes decomposition of CaF_2 and desorption of fluorine atoms occur.[3,4] The streaky RHEED image, corresponding to a flat or a stepped surfaces, changed to the spotty or halo RHEED images, indicating destruction of surface crystal order during the RHEED observation.

These phenomena disturb the analysis on ionic crystal surfaces by use of RHEED. The gradual change in lattice constant is a feature of the heteroepitaxy of alkali halide, one of the most typical ionic crystals.[5] In the case of the RHEED observation the velocity of change in surface lattice constant sometimes depends on the intensity of monitoring electron current. As the electron current increases, the change in lattice constant is delayed. This is probably because impinging electrons decomposed the growing film and the deposition amount was reduced. In the previous works, the RHEED image was observed intermittently to reduce the effect of probing electrons.[2,5] In the present work, we used a micro-channel plate (MCP) to enhance the RHEED image, expecting a large amount of reduction in probing electron current.

II. MCP-RHEED SYSTEM

Figure 1 shows the experimental setup of MCP–RHEED system. MCP (Hamamatsu Co. Ltd., F2226), which is mounted on an ICF 203 flange, is placed in front of a fluorescent screen plate. The effective diameter of the MCP is 77 mm. The diffracted electron beam from the specimen enters the MCP front and is multiplied by electric field in the channel tubes. The gain depends on the voltage applied to MCP, V_{MCP}, and it reaches four to five orders of magnitude for V_{MCP} of 1 kV. The enhanced diffraction beam was accelerated to the fluorescent screen by the electric field between the MCP and the screen. The voltage (V_s) was adjusted between 1 and 4 kV to optimize the contrast of the image. It was typically 3.5 kV in the present experiment.

We measured the sample current necessary to get the RHEED image of good quality with or without MCP. We obtained the relation between the filament current (I_f) of the RHEED gun and the probing electron current (I_p), which was measured by a Faraday cup placed at the specimen point. Without MCP the typical I_f ranges from 4.4 A to 4.9 A, corresponding to I_p of 100 nA to 1 μA. With MCP, the RHEED image could be obtained even for I_f of 3.1 A to 3.4 A. In this case I_p is estimated to range from 1 pA to 20 pA, which is smaller by four orders of magnitude than the case without MCP. The extreme decrease in probing electron current prevents the electrolysis of ionic crystal surfaces during the RHEED observation.

III. RESULTS

With MCP, several experiments on the heteroepitaxial growth of alkali halide have been done. In the following,

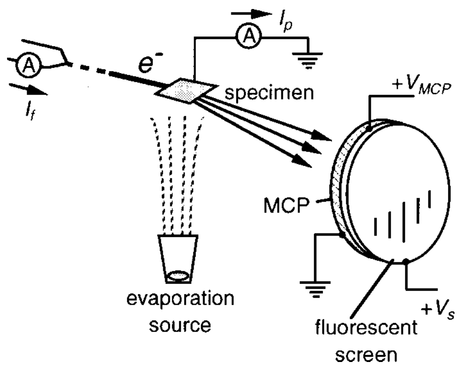

FIG. 1. A schematic view of the MCP-RHEED system.

three typical examples will be described to show the effectiveness of reduction of probing electron beam in the RHEED analysis. First, continuous RHEED observation could be achieved during the growth of NaBr on KCl. The left column of Fig. 2 shows the time evolution of the RHEED rocking image. In the right column the line profiles are shown, which correspond to the dashed positions in the left image. There are three lines corresponding to three diffraction rods; [1, 1], [0, 0], and $[\bar{1},\bar{1}]$. From the top to the bottom the line interval increases, indicating the decrease in surface lattice constant, because the interval between RHEED streaks is inversely proportional to the lattice spacing. Without MCP, the streak interval showed no change during continuous observation. Instead the streak became broad, suggesting the lattice disorder in the surface region.

Although the change in streak interval is observed in Fig. 2, it is more favorable to observe the RHEED image intermittently even with use of MCP. The effect of electron

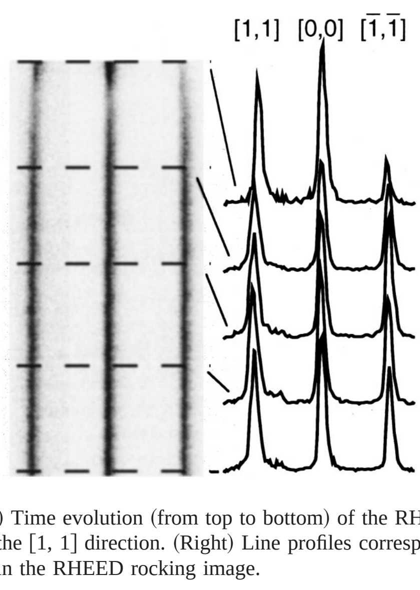

FIG. 2. (Left) Time evolution (from top to bottom) of the RHEED rocking image along the [1, 1] direction. (Right) Line profiles corresponding to the dashed lines in the RHEED rocking image.

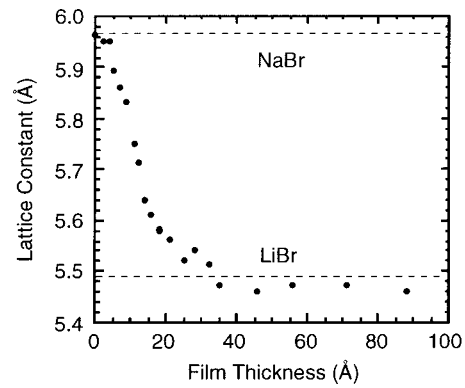

FIG. 3. Change in lattice constant during growth of LiBr on NaBr.

beam should be reduced as much as possible for precise discussion on the growth mechanism of ionic crystals. Figure 3 shows the change in lattice constant of LiBr during the growth on a NaBr substrate, which was obtained by the intermittent observation of the RHEED. The change in surface lattice constant could be observed precisely every 1 Å growth of LiBr. The result indicates that the surface lattice constant of LiBr relaxes to the bulk value over 30 Å. With this system the growth feature of alkali halide and other highly insulating materials can be investigated. This will be reported elsewhere.

The third example is observation of growth on a LiF substrate. LiF has the largest bandgap (13.6 eV) among all the solids. It is highly insulating such that the RHEED observation could not be possible for the temperatures lower than 250 °C. In the case of growth of alkali halide, this substrate temperature (T_s) is too high for the growth to occur. Moreover the LiF substrate itself partially evaporates at 250 °C. In the present observation with MCP, the RHEED image could be observed for T_s of 120 °C, for which the growth of alkali halide proceeds.

IV. DISCUSSION

In summary, use of MCP in the RHEED observation system enabled reduction of probing electron beam by four orders of magnitude. This helps the application of RHEED to insulating materials. It has been proved that the surface and film growth of ionic crystals were observed with less electron-induced damage and even the highly insulating surface was observed for the lower temperature.

[1] M. H. Yang and C. P. Flynn, Phys. Rev. B **41**, 8500 (1990).
[2] K. Saiki, Y. Nakamura, and A. Koma, Surf. Sci. **250**, 27 (1991).
[3] C. L. Strecker, W. E. Moddeman, and J. T. Grant, J. Appl. Phys. **52**, 6921 (1981).
[4] K. Saiki, Y. Sato, K. Ando, and A. Koma, Surf. Sci. **192**, 1 (1987).
[5] K. Saiki, Appl. Surf. Sci. **113/114**, 9 (1997).