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Kinetic Monte Carlo simulation of RHEED from $BaTiO_3$ thin films

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ABSTRACT

We proposed an energy-dependent kinetic Monte Carlo approach to simulate the reflection high-energy electron diffraction intensity during the three-dimensional growth of $BaTiO_3$ thin film using pulsed laser deposition. In the simulation, we employed the Born–Mayer–Huggins potential and assumed the overhanging of atoms in the deposition and diffusion process. The factors considered include the incident kinetic energy, laser repetition rate and mean deposition rate. By changing the values relative to every factor, the effects on the reflection high-energy electron diffraction (RHEED) intensity were studied. Being in good agreement with the experimental observations, the results provide an understanding on the evolution of the morphology about $BaTiO_3$ thin film and a basic exploration of the epitaxial growth process of ionic oxides with perovskite-type structures.

1. Introduction

Ferroelectric thin films have attracted much attention for the potential applications because of some outstanding characteristics [1]. $BaTiO_3$ thin film with perovskite structure is a typical ferroelectric thin film with high dielectric constants and high electro-optic coefficients [2]. $BaTiO_3$ thin films have been fabricated by various deposition techniques such as pulsed laser deposition (PLD) [3]. In practical applications, the ability to control the process of the $BaTiO_3$ thin films growth down to the atomic level is an obvious prerequisite. However, the understanding of the growth mechanism of oxide materials is incomplete and thus of fundamental interest. Reflection high-energy electron diffraction (RHEED) is a technique widely used to study epitaxial growth of a variety of materials. The schematic of RHEED is shown in Fig. 1. In RHEED, a high-energy electron beam is directed towards the sample at extreme grazing incidence. Electrons scattered through small angles sample only the top 1–2 atomic layers of the crystal under these conditions, therefore it is suitable to measure the structure of the sample and can detect the growth of thin films [4]. The RHEED oscillation is due to a cyclic transition from a smooth to a rough surface, and vice versa; so the RHEED oscillation is currently used to check the two-dimensional (2D) growth mode.

Up to now, a few experimental reports have been obtained on the RHEED intensity of ferroelectric thin films [5,6], whereas there are some difficulties on detecting surface due to the complicated perovskite structure. Therefore, the simulation of RHEED from thin films is necessary. In Ref. [7], it was shown by the authors that the RHEED intensity depends inversely on the surface step density, and this fact was used to simulate the RHEED intensities by Monte Carlo methods [7,8], which can give the correct experimental evidence for the process of thin film growth. Nevertheless, these were taken care in the metal and semiconductor thin films. Based on the previous work [9], of developing a 2D model and algorithm about growth process via PLD, here we develop a three-dimensional (3D) model to simulate the RHEED intensity of $BaTiO_3$ thin films at different parameters. Through the simulation of RHEED, the detailed evolution of surface morphology can be studied well, so that it may provide some theoretical basis for the preparation of the ABO_3 perovskite thin film.

2. Three-dimensional model

Fig. 2(a) shows a molecular cell with the perovskite structure; there are O, Ti, and Ba atoms in the face center, bulk center, and apex of the cell, respectively. The cubic lattice space for the

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movement of atoms in the KMC simulation is determined by the number of cubic sites of $L \times L \times H$, where L and H are the length (also the width) and the height of this lattice space. L is 100 and H is related to the number of incident atoms. It is assumed that the length of one side of each of the cubic sites as illustrated in Fig. 2(b) is 0.2 nm. Each site can hold an atom. The periodic boundary condition is adopted in the horizontal plane. The n atoms from each pulse are assumed to be instantaneously deposited at random sites on the $L \times L$ surface area. In order to describe the deposition process, the coverage is determined by $\theta = N/L^2$. The number of total incident atoms N is set as 50,000, and the maximum of total coverage is 5.0 ML. The model of fabrication process of $BaTiO_3$ thin films is considered as described in the following.

2.1. The deposition of atoms

In PLD, atoms in the target material are ablated by a pulsed laser, and then the energetic atoms are deposited onto the substrate surface. During a pulse, the atoms are deposited at randomly chosen positions on the surface of substrate or thin film at an instantaneous flux F_i, while no atom is deposited during the time interval between two pulses. The average flux of incident atoms is $F_{aver} = t_p \times f \times F_i$, with t_p and f representing the pulse duration of incident flux and laser repetition rate, respectively. This kind of deposition atoms is determined according to the ratio of Ba:Ti:O = 1:1:3 (as the atom ratio of $BaTiO_3$) by a stochastic number generated by computer. Such considerations are based on the fact that the kind of an atom deposited at one moment is randomly selected, but the ratio of atoms is 1:1:3 for the $BaTiO_3$ thin film. The incident kinetic energy is simplified as uniform for all of the deposited atoms. Considering the characteristic of the perovskite structure, the overhanging atoms can exist during the process of deposition. For the deposition of Ba (O) atom to the surface (plotted by the circle of dotted line) it is permitted to overhang on its two first-nearest-neighbor O (Ba) atoms, but Ti atom is not, as illustrated in Fig. 3(a).

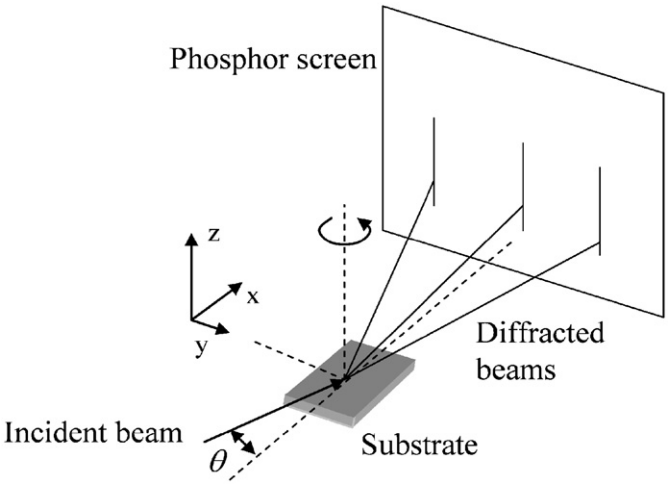

Fig. 1. Schematic view of the RHEED geometry.

2.2. The diffusion of adatoms

The diffusion probability P_i of an atom in the direction i can be calculated by the formulae

$$P_i = v_i / \sum_i v_i, \tag{1}$$

where v_i is the hopping frequency in the direction i which depends on the active energy and can be determined by [10]

$$v_i = v_0 \exp(-\Delta E/k_B T), \tag{2}$$

where v_0, k_B and T are, respectively, the attempt frequency (assumed to be 1.0×10^{12} Hz [11]), Boltzmann's constant, and substrate temperature. It is supposed that an atom at (x_0, y_0, z_0) can only diffuse to four nearest neighboring lattice sites such as (x_0+1, y_0, z_0), (x_0-1, y_0, z_0), (x_0, y_0+1, z_0), (x_0, y_0-1, z_0) in the same plane (Fig. 3(c)), and (x_0, y_0, z_0-1) can also be allowed to diffuse if the site is not occupied in the substrate (Fig. 3(d)). If there are atoms in the sites, the diffusion is prohibited to the sites. In Fig. 3(b), the Ba (O) atom plotted by the circle of dotted line presents the position before diffusion and the overhanging Ba (O) atom is permitted during the diffusion. The diffusion time Δt for one step can be calculated by

$$\Delta t = 1 / \sum_i v_i. \tag{3}$$

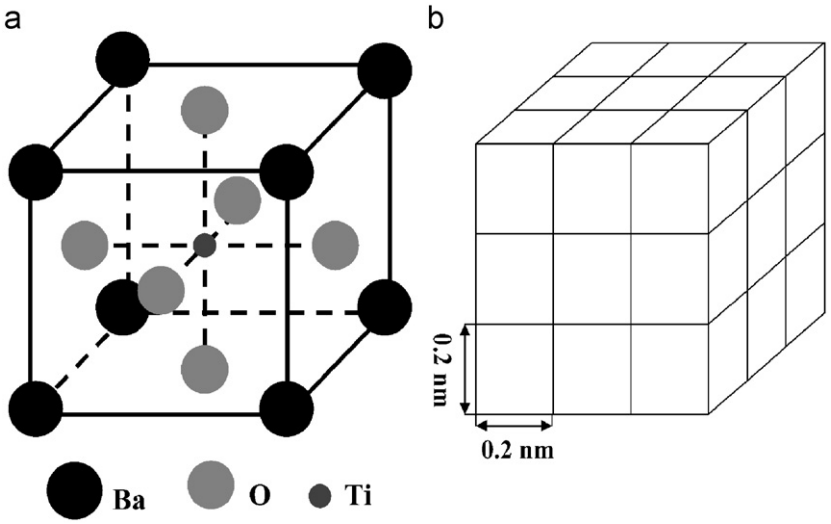

Fig. 2. (a) $BaTiO_3$ perovskite structure and (b) cubic lattice space.