Broadening of Cr nanostructures in laser-focused atomic deposition*

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This paper presents the experimental progress of laser-focused Cr atomic deposition and the experimental condition. The result is an accurate array of lines with a periodicity of 212.8±0.2 nm and mean full-width at half maximum as approximately 95 nm. Surface growth in laser-focused Cr atomic deposition is modeled and studied by kinetic Monte Carlo simulation including two events: the one is that atom trajectories in laser standing wave are simulated with the semiclassical equations of motion to obtain the deposition position; the other is that adatom diffuses by considering two major diffusion processes, namely, terrace diffusion and step-edge descending. Comparing with experimental results (Anderson W R, Bradley C C, McClelland J J and Celotta R J 1999 *Phys. Rev.* A **59** 2476), it finds that the simulated trend of dependence on feature width is in agreement with the power of standing wave, the other two simulated trends are the same in the initial stage. These results demonstrate that some surface diffusion processes play important role in feature width broadening. Numerical result also shows that high incoming beam flux of atoms deposited redounds to decrease the distance between adatoms which can diffuse to minimize the feature width and enhance the contrast.

Keywords: atom optics, laser-focused atomic deposition, kinetic Monte Carlo simulation, surface growth

PACC: 3280, 3380P, 4250V

1. Introduction

Nowadays many new approaches can be used in the fabrication of nanometer-size structure. Laser-focused atomic deposition is a new technique for nanofabrication in which atoms are deposited onto a substrate through a near-resonant laser standing wave and formed into parallel lines or dots onto a substrate. In 1992, for the first time Timp *et al.*[1] demonstrated that Na atoms could be focused into a grating-like structure by laser-focused atomic deposition experiment. One year later McClelland *et al.* reported the Cr atomic deposition[2] and analysed the difference between the line shape of the model prediction and the experimental result, concluding that this could be attributed to the surface diffusion as one of the factors. For this reason, their group had advanced greatly in laser-focused Cr atomic deposition. Experimental works by McClelland,[3] Behringer[4] and Anderson[5] had suggested that surface diffusion was a significant part in feature width broadening. For example, Anderson *et al.*[5] found that the structures deposited were always 20–30 nm wider than the model prediction. They concluded that the feature width broadening must be caused by a diffusion process on the substrate. To investigate the diffusion effect, Jurdik *et al.*[6] performed Monte Carlo (MC) simulation of deposition. In simulation, three models were studied, including ballistic deposition, ballistic deposition with relaxation and Tamborenea and Das Sarma model. Bradley *et al.*[7] applied the continuous approach relied on a solution to a partial differential growth equation and found the theory of the impact cascade diffusion model. But this theory did not explain the experimental phenomenon.[4] Zhong *et al.*[8] introduced terrace diffusion and step-edge descending processes and kinetic Monte Carlo (KMC) method to simulate the surface diffusion of laser-focused atomic deposition. Because the atomic flux density profile was directly assumed to be a Gaussian distribution and semiclassical equation of motion was not considered during the deposition event, the model could not explain the dependence on the feature width and power of standing wave. Sligte *et al.*[9,10] also used a KMC approach taking into account the Ehrlich–Schwoebel (ES) barrier and pollution from the residual gas in the vacuum system. Although the models, especially the pollutant adatom hypothesis, could explain the dependen-

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cies of the structure width on low temperature (less than 280 K) and vacuum conditions,[4,5] the atomic flux density profile in simulation is not the experimental result or the model prediction but a Lorentz distribution.

For the purpose of minimizing feature width, we attempt to relate some experimental results with surface growth in laser-focused Cr atomic deposition. We take into account the effect of some surface diffusion processes which might be limited by terrace diffusion and step-edge descending.[11,12] The step-edge diffusion is well known in the molecular beam epitaxy (MBE) and is called the ES barrier.[13] In traditional MBE growth, the above two diffusion processes[11] are of the most important function in controlling the morphological evolution of thin films.

The remainder of this article will be dedicated to our experimental result and numerical trends of feature width in laser-focused Cr atomic deposition. We begin by reporting our experimental result and some conditions in laser-focused Cr atomic deposition in Section 2, and will describe the surface growth model including terrace diffusion and step-edge descending in Section 3. Then, we show the simulated result of the model in Section 4 and conclude in Section 5.

2. Experiment

The ^{52}Cr atom is focused and deposited onto silicon substrate under the field of one-dimensional laser standing wave. The experimental scheme is referred to Ref. [14] in detail. Briefly, a beam of Cr atoms is produced by a high-temperature atomic oven at 1650 °C corresponding to a most-probable mean longitudinal velocity (V_Z) of 957 m/s. The atom beam is first pre-collimated with a mechanical aperture and then collimated with one-dimensional Doppler laser cooling. Finally, the atom beam is focused by standing wave and deposited on silicon substrate. Frequency stabilization technology based on laser-induced fluorescence[15] can obtain good laser source to cool or focus Cr atom. Wavelength of laser standing wave (λ) of 425.55 nm is chosen in respect that ^{52}Cr atomic resonance transition ($^7S_3-^7P_4^0$) is just 425.55 nm in vacuum. The $1/e^2$ intensity radius of the Gaussian beam (w_0) is 100 μm. The standing wave beam is blueshifted $\Delta/2\pi = 500$ MHz by an acoustic optic modulator. The total power of laser is 180 mW, including laser frequency stabilization of 24 mW, laser cooling of 31 mW and laser focusing of 13 mW. The centre of the incident Gaussian laser beam is separated from the substrate surface by one beam waist (w_0) for convenient operation. The room temperature is 20 °C. The deposition time is 62 min, and then the sample is examined by atomic force microscope (AFM) made in Veeco Instruments Inc. The sample AFM image showing the surface three-dimensional (3D) topography of a 5×5 μm area of deposition is displayed in Fig. 1(a). A lot of periodic parallel lines have been deposited

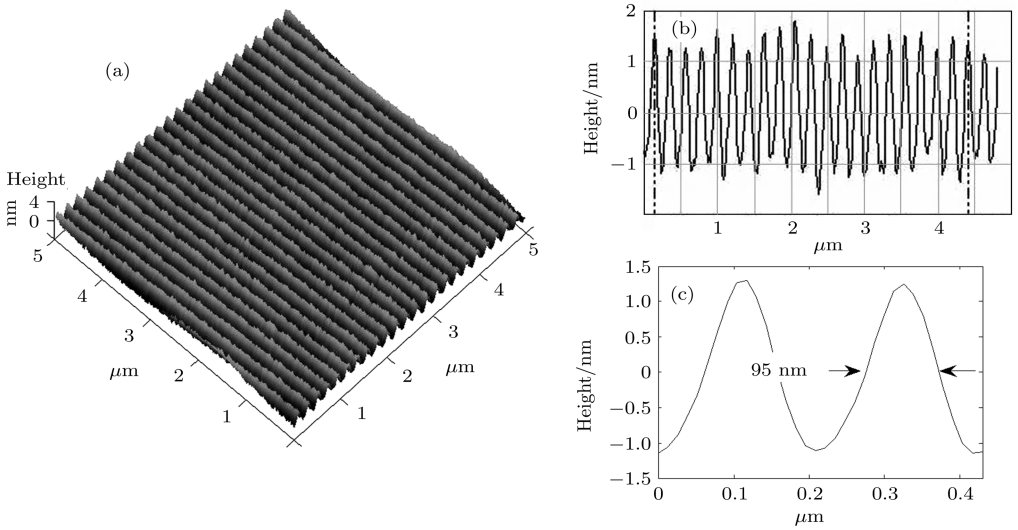


Fig. 1. (a) The 5×5 μm AFM 3D topography of lines formed by laser-focused Cr atomic deposition. (b) A linescan taken from (a) perpendicular to parallel line direction. (c) An averaged linescan carried out over a distance of 2.35 μm along Cr line direction. The average FWHM of the two peaks in (c) is approximately 95 nm and the peak-to-valley height is about 2.5 nm.

evidently on the silicon substrate. According to the datasheet of 2D PSD, the mean value of periodicity of Cr lines is 212.8±0.2 nm which is exactly equal to half of the laser wavelength $\lambda/2$. Figure 1(b) extracted from data of Fig. 1(a) perpendicular to parallel line direction is selected to plot the local detail. It shows that there is a nice periodic line shape. An average linescan carried out[5] along the Cr lines of Fig. 1(a) over a distance of approximately 2.35 μm has been measured and averaged. The feature width on the sample shown in Fig. 1(c) has an average full width at half maximum (FWHM) of approximately 95 nm and a peak-to-valley height of about 2.5 nm.

Although periodic Cr atom lines were deposited on silicon substrate and mean periodicity of $\lambda/2$ was accurately obtained, the feature width presented by FWHM and the peak-to-valley is extremely less than satisfactory. Next experimental work focuses on minimizing the feature width and enhancing the peak-to-valley by means of all possible parameters.

3. Model of surface growth

In fact, the formation of nano-periodicity is not a simple hit-and-stick process but perhaps exists in thermal migration or diffusion process. Understanding the diffusion effect happened during or after deposition is therefore of vital importance for predicting and controlling the structure feature size.

Our model is a solid-on-solid model where the substrate is represented by a simple cubic lattice. Periodic boundary conditions are applied to the lattice. The model is limited to one dimension for limiting the complexity of the programme code and reducing the running time. In the simulations, we create a list of all the possible events, including deposition event and diffusion event mentioned later, and one event is selected randomly at a time according to a weighted probability. At each MC step (time), the probability P_i of a given event being selected is given by:[16]

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

where R_i is the rate of an event, and the summation is over all possible events. Thus at each MC step, an event is chosen to occur, i.e., an atom either moves from one site to another site or deposits. Iteration of these processes simulates the nature in laser-focused Cr atomic deposition with diffusion process. The KMC algorithm[17] is used repeatedly until the total amount of atoms deposited satisfies our request.

In the deposition event, one atom randomly enters standing wave and then the position of deposition is calculated with semiclassical equation of motion. Our basic approach based on the semiclassical equation of motion has been described in detail.[18] For advancing calculation accuracy, we introduce Adams–Bashforth–Moulton algorithm[19] instead of four-step Runge–Kutta algorithm.[18] Essentially, a lots of classical trajectories through the 'atom lens' formed by standing wave are calculated for random initial conditions of position, velocity and incident angle. Each trajectory is assigned a probability according to a uniform spatial distribution, a thermal longitudinal velocity distribution and a transverse velocity distribution. The deposition rate is:[17]

$$R_0 = FN, \tag{2}$$

where F is the incoming beam flux of atoms deposited and N is the number of sites for deposition.

In the adatom diffusion event, there are two processes including terrace diffusion and step edge descending. Once adatom is on the terrace, the adatom is able to hop from site to site if the probability of hopping is larger than a random number. We assume all hopping processes are likely equally, provided that the adatom can move without reducing their number of nearest neighbours. The hopping frequency of terrace diffusion or step-edge descending is assumed constant respectively, and given by the Arrhenius equation[16]

$$R_i = \Gamma_0 \, \mathrm{e}^{-E_\mathrm{A}/k_\mathrm{B}T}, \tag{3}$$

where Γ_0 is a prefactor related to the vibrational frequency, typically of the order of 10^{12} Hz, k_B is the Boltzmann constant, and T is the absolute temperature of substrate, E_A is the diffusion activation enthalpy. Therefore, the hopping frequency of terrace diffusion and step-edge descending are respectively[10]

$$\begin{aligned} R_\mathrm{T} &= \Gamma_0 \, \mathrm{e}^{-E_\mathrm{T}/k_\mathrm{B}T}, \\ R_\mathrm{ES} &= \Gamma_0 \, \mathrm{e}^{-(E_\mathrm{T}+E_\mathrm{ES})/k_\mathrm{B}T}. \end{aligned} \tag{4}$$

In surface physics, E_T and E_ES are different according to different material, different lattice plane, and so on. In general, E_T is larger than E_ES in the same case. In other words, terrace diffusion is easier to happen than step-edge descending in the diffusion event.

Actually, we note that adatom diffusion is different between Si–Cr and Cr–Cr during growth, i.e., $E_{\rm T}$ and $E_{\rm ES}$ are different between different substrates. For simplicity, we just consider the adatom diffusion in the case of Cr–Cr.

4. Result of KMC simulations: discussion

The aim of the simulation presented in this paper is to model some feature width trends referred to some experiments in laser-focused Cr atomic deposition. We can verify some understanding of the feature width broadening by comparing the trends predicted by the model with those of experiments.

In the simulation, the grid spacing is assumed to be equal to the lattice constant for Cr. The grid size to the period of the incoming beam flux distribution is equal to 212 nm for Cr. This corresponds to 739 sites for a lattice constant of 0.287 nm. So the number of sites for deposition (N) is 739.[10] In the deposition event, we assumed $w_0 = 100$ μm, $\Delta/2\pi = 250$ MHz, $V_Z = 957$ m/s, $I_{\rm sat} = 85$ W/m^2, $\gamma/2\pi = 5$ MHz, $P = 3.93$ mW. The centre of the incident Gaussian laser profile intercepts the substrate surface, i.e., the position of deposition (z) is 0 mm. Transverse velocity distribution and longitudinal velocity distribution are not considered just for simplifying the model. In the diffusion event, the terrace diffusion barrier ($V_{\rm T}$) is 0.34 eV and the step edge descending barrier ($V_{\rm ES}$) is 0.05 eV. The temperature of substrate (T) is 300 K and the incoming beam flux of atom deposited (F) is 0.05 ML/s. All the parameters above are fixed if not stated again.

Before investigating the dependencies of this model, first, we look at the predicted difference of feature width. Figure 2 displays laser-focused Cr atomic deposition without diffusion (dot and dash lines) and deposition with diffusion (full line) when the total amount of atoms deposited is 5000. Especially, the central part is amplified in the top right corner of Fig. 2 for discriminating. It shows that the deposition with an FWHM of less than 1.5 nm (dot and dash lines) is transformed into a structure with FWHM of 36.5 nm (full line). As can be seen, the resulting feature width becomes broader and lower than that without diffusion. Obviously, the terrace diffusion and step-edge descending plays a significant role in feature width broadening. Here, we select a power (P) of standing wave of 3.93 mW for the reason that both good contrast[20] and sharp focusing are achieved for the paraxial rays of the perfectly collimated atomic beam by using a numerical result of the formula[21] with the above parameter values.

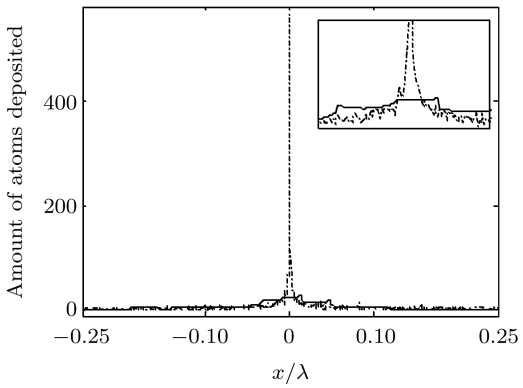


Fig. 2. Sample simulation results. Dot and dash lines: deposition without diffusion. Full line: deposition with diffusion. The central part is amplified in the top right corner.

In order to minimize the feature width, it is crucial that the standing wave focuses the atom beam into a pattern with high contrast and narrow linewidth.[4] Figure 3 shows the feature width as a function of standing wave power in the case of the total amount of atoms deposited of 5000. We find that as the power of standing wave increases it shows a decrease in the FWHM, after which there is an increase just like the experimental trend of Fig. 5(a).[5] So the power of standing wave using in the experiment needs to be optimized and is sometimes different from that only predicted by radiative dipole forces because of surface growth.

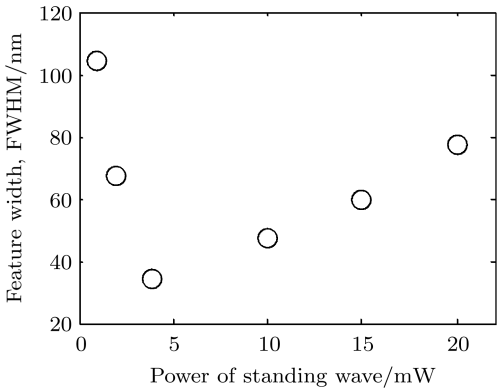


Fig. 3. Feature width as a function of power of standing wave.

According to Fig. 6,[5] the trend is simulated by Fig. 4 shown the feature width as a function of amount

of atoms deposited. Comparing Fig. 4 with Fig. 6,[5] we find that the trend is in excellent agreement with the decrease in the initial stage. But as the total amount of atoms deposited increases, the trend of Fig. 4 disagrees with the trend of Fig. 6[5] and the FWHM simulated is nearly equal to the one without diffusion. This demonstrates that the feature width broadening phenomenon induced by the terrace diffusion and step-edge descending is not strong any more. It indicates that other processes of diffusion may contribute significantly to the feature width broadening except for the above two. However, there exists the feature width broadening limited by terrace diffusion and step-edge descending in the initial stage.

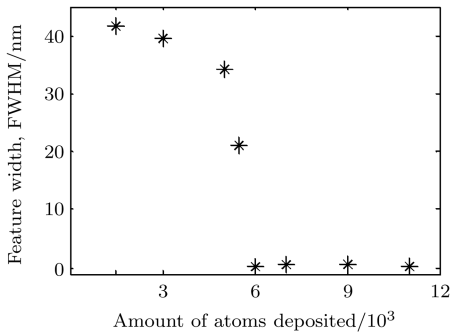


Fig. 4. Feature width as a function of amount of atoms deposited.

Figure 5 shows the dependence on the change in feature width and overlayer thickness presented by amount of atoms deposited. This is a very important characteristics to confirm the effect of adatom diffusion. To perform this simulation, laser-focused Cr deposition is modified by depositing a uniform layer of atoms (overlayer) on top of the patterned depositions (base layer). The deposition contains the features with nominal dimensions of 41.5-nm FWHM. Figure 5 displays that there is an increase as the amount of atoms deposited increases. The trend of feature width is the same when the amount of atoms deposited is less than 1000 in comparison with the trend of Fig. 7.[5] But as the thickness (amount of atoms deposited) increases, the numerical trend cannot explain the experimental trend[5] just as figure 4 reveals. Therefore, other diffusion mechanisms may exert simultaneously influence and should be considered in surface growth model. But both figures 5 and 7[5] demonstrate that the formation of nano-periodicity is not a simple hit-and-stick process since the feature width broadening is not equal to 0 nm as the overlayer increases.

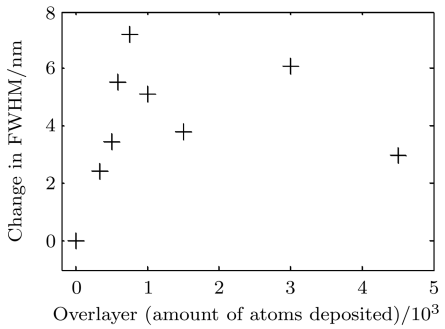


Fig. 5. Feature width as a function of overlayer (amount of atoms deposited).

In growth of film, the incoming beam flux of deposited atoms (F) is one of the most pivotal factors. Figure 6 shows the feature width and contrast as a function of the incoming beam flux of deposited atoms (F). As can be seen, the feature width can be reduced and the contrast can be enhanced by increasing the incoming beam flux in the case that the total amount of atoms deposited is 1500. It is reasonable that high incoming beam flux can decrease the distance between adatoms which diffuse so that the feature width broadening is limited. Therefore, increasing the incoming beam flux of atoms deposited redound to minimizing the feature width. At the same time, increasing the detuning in standing wave is also necessary to enhance the atomic focusing of laser standing wave.[4]

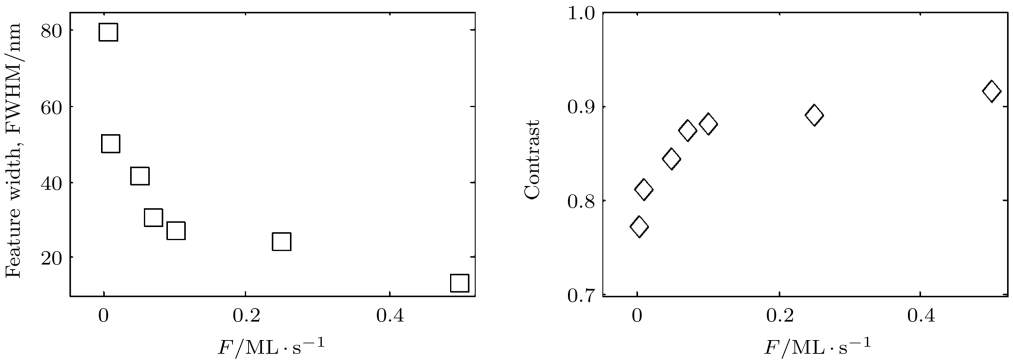


Fig. 6. Feature width and contrast as a function of the incoming beam flux of deposited atoms (F).

The temperature of substrate (T) is also one of the most important factors during the growth. The dependence on the temperature of substrate and the feature width had been investigated by some authors[8,10] and used to compare the experimental phenomenon.[5] But their models cannot explain the experimental result[5] effectually. So it is not considered by us for lack of more diffusion mechanisms at present.

5. Conclusion

We have presented our experimental progress in laser-focused Cr atomic deposition. The periodicity is 212.8±0.2 nm which is exactly equal to $\lambda/2$ and the mean FWHM is about 95 nm. Then, we have modeled the surface growth by considering two kinds of atomic diffusion processes, namely terrace diffusion and step-edge descending. We have performed KMC simulation of two events including deposition event and diffusion event. The numerical result shows that the trend of feature width as a function of standing wave power is in agreement with the experimental trend[5] and other two trends also match those[5] in part. These demonstrate that terrace diffusion and step edge descending are crucial parts in the feature width broadening at least in the initial stage of surface growth. Otherwise, other diffusion mechanisms may exert an influence on the surface growth and should be considered. Overall, the surface growth model could be further refined by the inclusion of other diffusion processes, such as the impact cascade diffusion.[7] This will be the future object of surface growth in laser-focused Cr atomic deposition.

To improve experimental result, a lot of factors should be optimized. From surface growth viewpoint, the incoming beam flux of deposited atoms is one of the main factors, and then we can attempt to increase incoming beam flux of atoms deposited. In the meantime, appropriate power of standing wave, the larger detuning in laser standing wave and lower temperature of substrate should be paid more attention.

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