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Superiority of Langmuir–Blodgett resist films in electron beam lithography as demonstrated by the backscattering yield

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Abstract

Applying Monte Carlo techniques, we have simulated the electron trajectories in a Langmuir–Blodgett (LB) polymethylmethacrylate (PMMA) resist and a Si substrate covered with a Cr film. A "refraction" model of high energy electrons in multilayer media is first used to describe the changes in scattering distance and scattering angle of an electron while it passes from one layer into another layer. In a Monte Carlo simulation, the trajectories of high energy electrons are calculated. The results of electron beam exposure experiments demonstrate the accuracy of the refraction model. The electron backscattering yields in PMMA LB resist and spin-cast resist have been calculated for various conditions. Finally, the superiorities of LB film resists in electron beam exposure lithography are discussed.

1. Introduction

Langmuir–Blodgett (LB) techniques enable transfer of a monolayer film from an air–liquid interface to a wafer [1]. The ultrathin, compact and uniform polymer films which are sensitive to electron beams and the deep UV prepared by LB techniques can be used as high resolution resists [2, 3]. As a kind of ultrathin resist, relative to conventional spin-cast resist films, the LB resist films have the advantages of low pinhole density, adequate etch resistance and ability to cover surface topography [4]. Furthermore, the LB resist films have other advantages in electron beam lithography. In this paper, applying Monte Carlo techniques, we simulated the electron trajectories and backscattering yields in LB and spin-cast polymethylmethacrylate (PMMA) resist and substrate. From the electron backscattering yields in LB and spin-cast resist films, we have evaluated the superiorities of LB resist films in electron beam exposure lithography.

2. Monte Carlo calculations

The basic computing model for the calculations of electron trajectories we adopted is similar to that used previously by other researchers [5–7]. However, in their calculations the changes in scattering distance and scattering angle or direction have been neglected while electrons pass from one layer into another. In fact, this assumption is not sufficiently accurate. We proposed an electron "refraction" model in multilayer media. In Fig. 1, we assume the present position of the electron in the first medium is A. Its energy is E_A. The scattering distance S_1, the scattering angle θ_1, and the scatter direction AO are calculated by the Monte Carlo method. If $S_1 > \overline{AO}$, the electron will pass into the second medium. If under the assumption that the electron were only in a single medium the same as the second medium, its distance and angle of scattering would be S_2 and θ_2 respectively as calculated by the same method. Now according to the consistency of scatter-free paths between two collisions, we have

$$\frac{\text{AO}}{S_1} + \frac{\text{OB}}{S_2} = 1$$

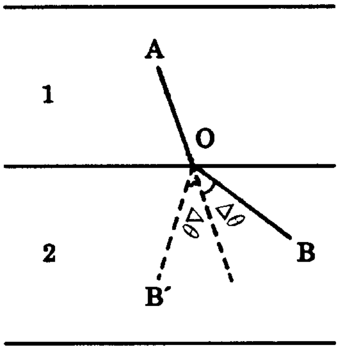

Fig. 1. A refraction model of a high energy electron in a multilayer medium.

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that is

$$OB = (1 - AO/S_1)S_2$$

Here, OB is the distance travelled by the electron in the second medium. The deviation angle $\Delta\theta$ between refraction direction and OA is $\theta_2 - \theta_1$. The next position B or B′ of the electron is determined by the sign of $\Delta\theta$. Correspondingly, the electron energy E_B at point B is

$$E_B = E_A - \int_{AO} \left(-\frac{dE}{dS}\right)_1 dS - \int_{OB} \left(-\frac{dE}{dS}\right)_2 dS$$

Here, $(dE/dS)_1$ and $(dE/dS)_2$ are the energy loss rates in the first layer and the second layer respectively. This can be obtained from the Bethe energy loss relation [8]. The model can easily be generalized to the situation where the electron passes through many layers within one scattering event.

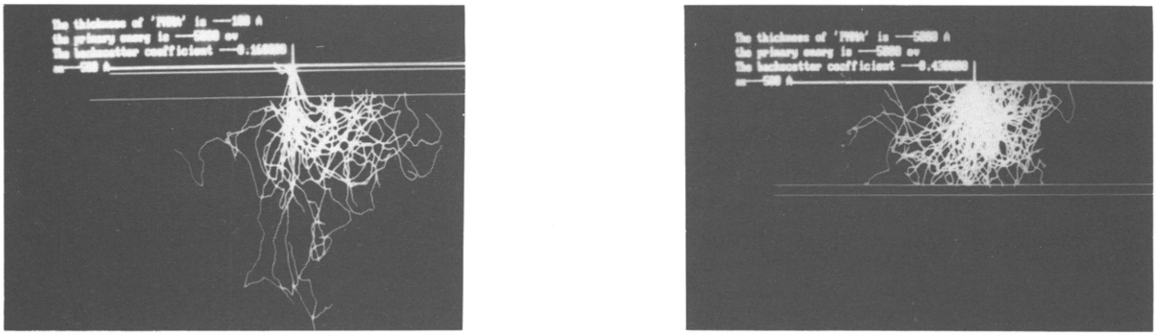
To calculate the electron trajectories in LB PMMA film resists, we must determine the density and the atom number per volume in LB PMMA film resists under the deposition conditions. The area per repeat unit under the deposition surface pressure can be acquired from the pressure–area isotherms of PMMA LB films [9]. Pease and co-workers have found that the thickness of a two-dimensional monolayer PMMA film on a subphase surface is 8.5 Å through ellipsometry [4]. Therefore we can easily calculate the required parameters in our Monte Carlo calculations. The calculations followed the trajectory of an electron until its energy decreased below 500 eV or it collided with a nucleus such that it was backscattered from the solid.

3. Results and discussion

Applying the described model and Monte Carlo method, we have simulated the electron scattering trajectories in an LB PMMA resist and Si substrate covered with Cr film. Figure 2 shows the simulated electron scattering trajectories in LB and spin-cast PMMA resist films. From Fig. 2, we see that the electrons with a 5 keV primitive energy have insufficient energy to penetrate the spin-cast films. As a result, at this low accelerating voltage, the spin-cast resist films will appear insufficiently exposed.

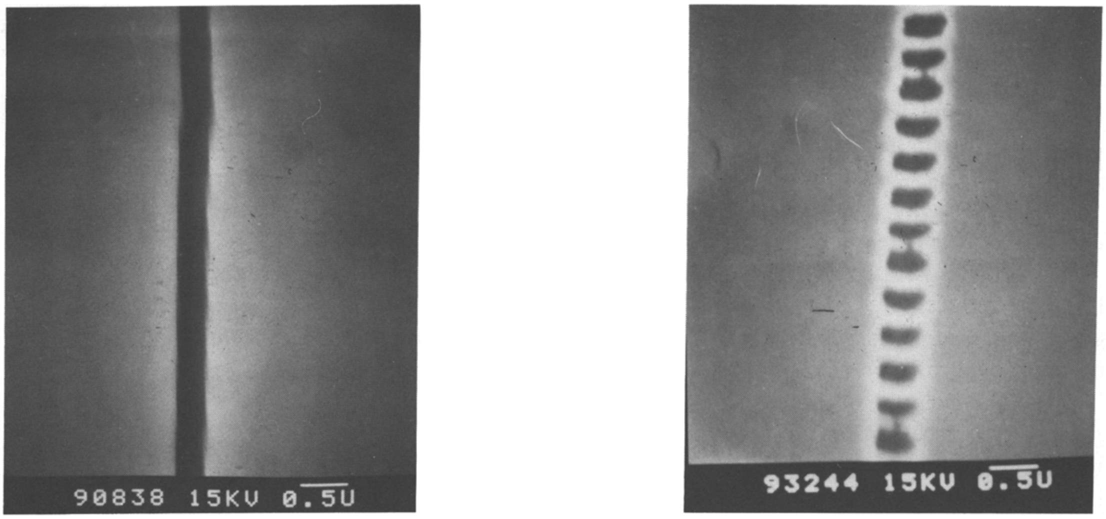
The results of electron beam lithography experiments on LB resist film and spin-cast resist film have verified the computed results. The electron beam exposure system in our study is a modified scanning electron microscope controlled by a computer. Two kinds of PMMA resist films have been fabricated. One was 12-layer (102 Å) PMMA LB film which was deposited on a silicon substrate evaporated with a 500 Å chrome film by continuous horizontal deposition techniques. Another film was spin-cast PMMA with a thickness of 0.5 μm on the same substrate which was prepared by spinning 65.6 mg ml^{-1} chlorobenzene solution at 8000 rev min^{-1} for 15 s. The system exposures were performed in point scanning mode, 5 kV accelerating voltage, 0.1 μm beam diameter, 1.5 pA beam current and a magnification of 1000 ×. The electron beam made a grating pattern with the scanning exposure controlled by a computer. After exposure, the samples were developed in a 1:3 solution of methyl isobutyl ketone:isopropyl alcohol for 30 s at 23 °C. The resists were post-baked at 80 °C for 40 min. After the developing process the samples were immersed in chrome etching solution to transfer the resist pattern to the Cr film. Figure 3 shows the etching results of the two resists observed by scanning electron microscopy (SEM). The exposure is apparently insufficient for the spin-cast resist films under the above-mentioned conditions. This is consistent with computed results. It demonstrates that the high energy electron refraction computational model is accurate.

In electron beam exposure lithography, the electron backscattering yield in resist and substrate is one of the key parameters because it causes the proximity effect



(a) (b)

Fig. 2. Simulated scatter trajectories of 100 electrons with 5 keV primitive energy: (a) 100 Å LB PMMA resist and Si substrate covered with 500 Å Cr film; (b) 5000 Å spin-cast resist and Si substrate covered with 500 Å Cr film.



(a) (b)

Fig. 3. The results of electron beam lithography carried out with a modified scanning electron microscope performed in point scanning mode, 5 kV accelerating voltage, 0.1 μm beam diameter, 1.5 pA beam current: (a) 102 Å LB PMMA resist and Si substrate covered with 500 Å Cr film; (b) 5000 Å spin-cast resist and Si substrate covered with 500 Å Cr film.

generated by electron scattering interactions. We have calculated the backscattering yields of electrons with different energies in LB PMMA resist and spin-cast films, as illustrated in Fig. 4. For accuracy in statistics, 2000 incident electrons are calculated for every condition.

From Fig. 4, relative to the usual spin-cast resist, there is a smaller backscattering yield in the LB resist under the same energy. In general, the factors decreasing the resolution because of backscattering in the electron beam exposure process are 10 times as large as the factors of forward scattering. A smaller backscattering yield reflects a smaller proximity effect. Therefore, preparation of the ultrathin LB resists is an effective technique for lessening the proximity effect and obtaining high lithographic resolution. In other words, in electron beam lithography, the use of LB resists will reduce electron scattering within the resists and thus make proximity effect correction schemes easier to implement.

Figure 5 is the curve of electron backscattering yield in an LB PMMA resist and Si substrate covered with a 1450 Å Cr film. For the usual spin-cast resists, the higher the beam energy is, the smaller the backscatter yield is generally. In other words, the simulation results demonstrate that the lower energy (up to 5 keV) cannot

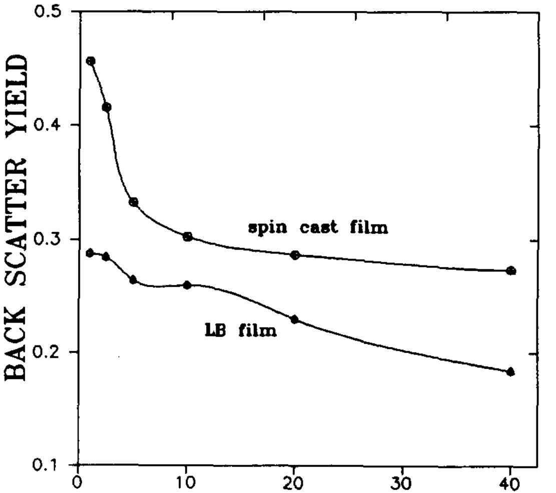


Fig. 4. The curves of backscattering yield of high energy electrons in 100 Å PMMA LB films and 5000 Å spin-cast films on silicon substrates.

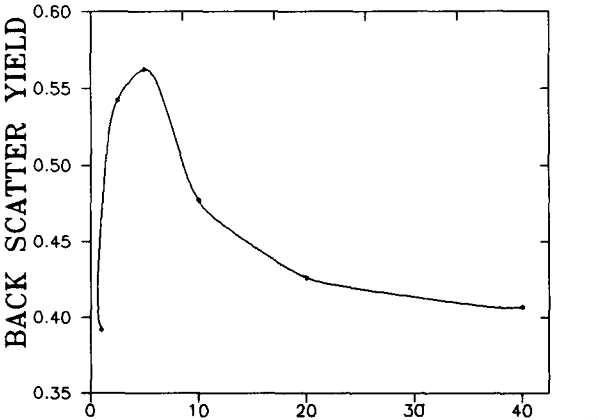


Fig. 5. The curve of electron backscattering yield in a 100 Å LB PMMA on an Si substrate covered with a 1450 Å Cr film.

let the electrons penetrate the 0.5 μm thick resist film to the substrate. However, this is not the case for the LB resists, especially for a high atomic number substrate. Figure 5 shows that there is a smaller backscatter yield whether for low beam energy or high beam energy for the Cr substrate. Comparing Figs. 4 and 5, we found that the backscattering yield is higher for the substrate which consists of 145 nm thick Cr over Si at the same primary beam energy. This demonstrates that the high backscattering yield can be reduced by using ultrathin resist films or by using higher beam energy or by using a substrate consisting of a lower atomic number material. High lithographic resolution can be obtained possibly whether a high or low accelerating voltage is adopted for LB resist films. This makes ultrahigh resolution lithography practical even if we adopt the relative simple electron beam exposure system, for example, modified SEM, etc. Moreover, because the scanning tunnelling microscope can generate electron beams with a very low voltage and extremely high current density, the LB resist will allow electron penetration when the scanning tunnelling microscope is used as an exposure tool. Because the scanning tunnelling microscope is capable of creating ultrahigh resolution patterns (better than 10 nm), it has become an important lithographic tool in nanofabrication in conjunction with the use of LB polymer films as resist materials.

4. Conclusions

We have demonstrated the improvements achievable using the refraction expression to model the electron scattering in multilayer films. The specially designed electron beam lithography experiments verified that this model provides an accurate description of the electron scattering in a multilayer resist–substrate system. From the Monte Carlo simulation results, the following conclusions are drawn.

(i) Under the same energy, the backscattering yields for LB resists are lower than those for the usual spin-cast resists.

(ii) There are lower backscattering yields in LB resist films whether a low or high electron beam energy is adopted.

(iii) In electron beam lithography, the high backscattering yield can be reduced by using ultrathin resist films or by using a high accelerating voltage or by using a low atomic number substrate.

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