Microelectronic Engineering 168 (2017) 41–44

Contents lists available at ScienceDirect

Microelectronic Engineering

journal homepage: www.elsevier.com/locate/mee

Research paper

On the spatial resolution limit of direct-write electron beam lithography

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ARTICLE INFO

Article history:
Received 14 July 2016
Received in revised form 11 October 2016
Accepted 18 October 2016
Available online 22 October 2016

Keywords:
Direct-write electron beam lithography
Scanning transmission electron microscopy
Electric field
Spatial resolution

ABSTRACT

The mechanism for direct-write electron-beam lithography in insulating resists is introduced in this letter, and it is based on damage by the induced electric field in transmission electron microscope. Under this mechanism, the direct-write EBL is electron dose-rate dependent, and there is a dose-rate threshold, below which the lithographic process does not operate, regardless of the total electron dose. The spatial resolution is determined by the strength of the induced electric field. In theory, the highest spatial resolution should be set by the dimension of the electron beam, and thus the EBL should be able to create nanostructures at the atomic scale. So far, the best resolution obtained was in the direct write of conductive nanochannels in $Li_4Ti_5O_{12}$, in which 1.5 nm isolated features and a 1.0–1.5 nm half-pitch array of nanochannels were achieved.

1. Introduction

Electron-beam lithography (EBL), a well-established high resolution patterning technique, has been widely used in nanotechnology and is the basis of much of the semiconductor device industry [1,3,9,22,26]. Understanding the limits of the spatial resolution in EBL is therefore very important in order to optimize the lithographic process. In brief, the resolution is determined by the following two processes: exposure (i.e. electron-resist interaction) and development in a developer [7,18]. For direct-write (one-step) EBL using self-developing resist, which is the focus of the current study, the latter does not apply, so that the resolution is only determined by the interaction range of the beam electrons with the resist. So far, it has been reported in all experiments that the lateral sizes of the lithographic features are always larger than the probe size of the beam [5]. The broadening due to elastic scattering of the beam electrons has not been considered to play a limiting role, instead, the limit of spatial resolution of EBL is believed to be set by secondary electrons (SEs) [14,20,27]. This SE model is supported by Monte Carlo simulations of SE trajectories, which calculate range of SEs in the resist. It was found that the broadening of the point-spread function (PSF) matches the extension of the SEs [14,20,27]. However, controversial results have also been reported. When Monte Carlo simulations track the energy deposited in the resist by SEs, instead of the range, the SEs have only slight, if any, effect on final resolution [4,5]. Besides SEs, delocalization of inelastic scattering of the beam electron has also been considered, and it has a similar interaction range as SEs [15]. Recently, attempts have been made to measure the EBL PSF directly using an aberration-corrected energy-filtered transmission electron microscopy (EFTEM) [17]. It was suggested that the volume plasmons should be more important than SEs in the limit of spatial resolution at the sub-10 nm scale. Even though the origins of these interactions are different, the delocalization effect is common in all the models. It seems that this delocalization effect provides a fundamental limit on efforts to achieve atomic resolution in EBL, although the finest electron beam can be focused within less than one tenth of a nanometer in diameter in the state-of-the-art electro-optical system [21].

Beside delocalization, all these existing models consider that EBL is an electron-dose dependent process, and thus different resists have different dose thresholds. Above the threshold (enough exposure), there are sufficient bond scission events to be developed by the developer [20] or atomic displacements to form a nanostructure in the direct-write EBL [17]. However, these models ignore important experimental evidence associated with EBL. In studies of hole-drilling in inorganic materials, it was discovered that the drilling process depends on the electron dose *rate* but not on total electron dose; prolonged exposure below the dose-rate threshold did not result in drilling [23]. In studies of nanofabrication using electron beams, it was found that the sizes of nanocylinders and nanowalls created by the electron beam is independent of specimen thickness [12,13]. Apparently, these observations violate the dose-dependent principle, and cannot be explained by these existing models. Recently, these phenomena have been interpreted by a revised mechanism of damage by the induced electric field (DIEF) [10,11]. In this study, we extend this mechanism to the EBL process, and especially discuss its impact to the limit of spatial resolution of direct-write EBL. Our results are based on experimental and theoretical analysis of thin, self-supporting films studied in scanning transmission electron microscope (STEM), free of backscattering. Although EBL has been studied for decades, the current study presents a very different view on the resolution limit from conventional beliefs. If this new model can be extended to more general situations, the atomic-level resolution could be achieved in EBL industry.

http://dx.doi.org/10.1016/j.mee.2016.10.016
0167-9317/

2. Experimental

For EBL in TEM or STEM, the specimen must be thin enough to allow incident electrons to pass through. Here there are two types of specimen, as illustrated in Fig. 1. One is equivalent to the self-supporting resist thin film (on the center in Fig. 1) and the other is equivalent to the resist thin film on a substrate (on the side). The specimen used for demonstration was $10Na_2O$-$20B_2O_3$-$70SiO_2$ (in mol%) glass, and was obtained by the conventional melt-and-quench method. TEM specimen was prepared by grinding the glass into powder in acetone, and picking them up using a Cu grid covered with a lacy carbon thin film. The EBL was carried out using the Cornell VG HB501 100 kV STEM, equipped with electron energy-loss spectroscopy (EELS). The probe size was about 0.25 nm in diameter, at which the probe current was about 0.4 nA.

3. Results and discussion

If the resist is insulating to electrons, the holes left by emissions of SEs and Auger electrons cannot be neutralized rapidly, resulting in accumulation of positive charges [2,8,10,11]. In the DIEF mechanism, the electric field is produced by these positive charges [10,11]. In the transmission geometry, for a thin self-supporting film, all the beam electrons traverse a thin slab of resist without depositing electrons inside it. In this case, the distribution of the induced electric field inside the resist is relatively simple. In most EBL systems, electron beams are highly focused, and their lateral dimensions are usually <0.5–1.0 nm, which is much smaller than the effective mean-free-path (MFP) of SEs and Auger electrons (e.g. >1.0 nm [24]). As illustrated in Fig. 2, most emitted electrons travel approximately perpendicular to the beam direction and scatter in a larger region around the beam column. This is equivalent to that incident electrons ionize the specimen into a positive inner core (nano-column) surrounded by a thick negative shell, and their volume ratio is r_0^2/r^2 (Fig. 2). Considering that $r_0 < 0.5$ nm and $r = 1$–100 nm [24], the volume of positively charged inner core is much smaller than the negatively charged shell, and thus the charge density of the positively charged electron-probed region is much higher than the negatively charged surrounding. Therefore, the exposed region can be considered as a positively charged nano-column, with the diameter of the electron probe and a length given by the resist's thickness for the case of a self-supporting thin film. Assuming that it is charged uniformly, the magnitude of the induced electric field for a given induced charge density ρ (Coulomb per length) can be simplified as [10]

$$|\mathbf{E}| = \frac{\rho}{2\pi\varepsilon_0\varepsilon_r R} \tag{1}$$

in which R is the shortest distance to the electron beam. Its direction points outward perpendicular to the beam. Thus the induced electric field has an approximately cylindrical symmetry around the beam [10]. This theory is supported by experimental observations of nanocylinders in silicate glasses [6,12,13] and nanochannels in $Li_4Ti_5O_{12}$ crystal [25] formed by a STEM probe. It should be noticed that for the sake of simplicity, we ignored the negative surrounding in deriving Eq. (1). This simplification does not affect the conclusion obtained, since including

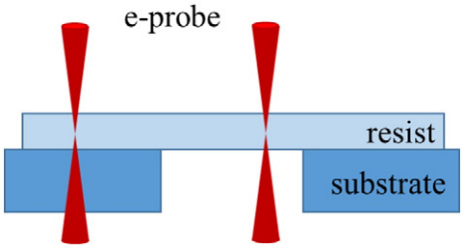

Fig. 1. Cartoon drawing showing the definition of self-supporting thin film.

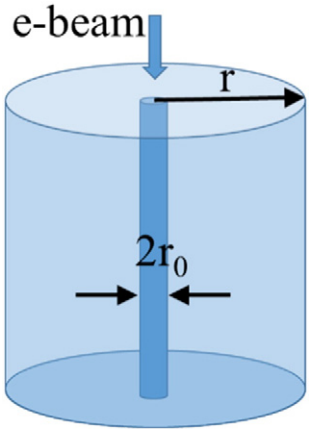

Fig. 2. Inner cylinder has net positive charges and larger outer one has net negative charges. r_0 is the radius of the electron probe, and r represents the range of emitted electrons.

these emitted electrons may further enhance the strength of the induced electric field, and thus lower the threshold beam current density. The range of r may not affect the resolution. This is because in this model the displacements of atoms are driven by the electric field, not by the ionization of secondary or plasmon electrons.

Interestingly, according to Eq. (1), the strength of the induced electric field is independent of the specimen thickness [10]. This is a unique characteristic of the DIEF mechanism in STEM, which distinguishes it from other dose-dependent mechanisms, such as knock-on and radiolytic processes. This thickness-independent characteristic has also been observed in previous direct-write EBL [6,12,13].

In resists, ionic bonds between anions and cations are polarized. Under a strong electric field, polar bonds can be ruptured, resulting in cation and/or anion displacements. To displace a bonded ion in a solid, the work done by the electric field on this ion as it moves from one site to the nearest available site must be equal to or larger than the activation energy U_a for ion migration, i.e.

$$W = \int_{site1}^{site2} q\mathbf{E}(\mathbf{r}) \cdot d\mathbf{r} \geq U_a \tag{2}$$

in which q is the electric charge of the ion. The distance d between the two nearest minimum energy sites for the ion should be in order of the nearest atomic distance, which is several angstroms. Assuming that $|\mathbf{E}(\mathbf{r})|$ does not vary significantly over such a small distance, the minimum work required for an atomic displacement can be simplified as

$$W_{min} = q\left|E_{ind}^{Th}\right|d = U_a \tag{3}$$

Therefore, the threshold strength $|E_{ind}^{Th}| = U_a/qd$, below which the lithographic process will not happen.

Fig. 3 shows some nanocylinders created by STEM probe in a Na borosilicate glass. The formation of the nanocylinder is due to the accumulation of mobile cations driven by the induced electric field [10–13]. Therefore, it is reasonable to consider that the strength of the electric field at the boundaries should be approximately equal to $|E_{ind}^{Th}|$. Na migration in silicate glasses has been extensively studied; in a glass containing 10 mol% Na, $E_a \approx 0.9$ eV [19]. The simplest approximation for the charge q of Na ion is to use its formal valence charge, i.e. $q = +1(e)$, where $e = 1.602 \times 10^{-19}$C (see S.I.). Here we set $d = 0.3$ nm, which is about the Na–Na distance in silicates. Inserting all these values in Eq. (3), we can estimate that $|E_{ind}^{Th}| = 3.0$ V/nm. This is the minimum electric field required in order to displace a Na ion in the $[SiO_4]$