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Beam damage by the induced electric field in transmission electron microscopy

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ABSTRACT

Electric fields can be induced by electron irradiation of insulating thin film materials. In this work, the electric fields under a broad beam illumination in transmission electron microscopy (TEM) are analyzed for insulating samples. Some damage phenomena observed can be interpreted by the mechanism of damage by the induced electric field (DIEF). For broad-beam illumination in an ultra-thin specimen, the electric field near the center of the illumination may not be strong, but at the periphery of the illumination the electric field can be significant. Therefore, damage may be easily observed in these regions rather than at the center of the illumination. For a beam which is broad compared to the specimen thickness, e.g. 100 ∼ 1000 nm, a strong electric field pointing inward into the specimen near the surface region may result in cation diffusion into the specimen and/or anion diffusion out to the surface region. Meanwhile, a strong electric field perpendicular to the beam direction near the edge of the illumination may attract anions into the illuminated region, but eject cations to the periphery. For a wedge-shaped specimen, the electric field points inward into thicker region, driving cations toward the thicker region, while attracting anions to the edge region. On the sharp edge, a strong electric field pointing outward may be responsible for the edge-smoothing effect observed in insulating materials.

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1. Introduction

Beam damage limits the experimental resolution of state-of-the-art transmission electron microscopy (TEM) (Egerton, 2013). Despite its importance, our understanding of damage mechanisms has not been as highly valued as it should be. Previously, most TEM studies have been trying to fit damage phenomena into the mechanism of either knock-on interaction, or radiolysis, or a combination of these two (Reimer, 1989; Williams and Barry, 1996). In brief, knock-on damage causes surface sputtering, mostly on the exit surface (Crozier et al., 1990). Radiolysis results in chemical-bond breakage, and thus produces point defects (e.g. Frenkel pairs) (Hobbs, 1990). However, beam damage observed in TEM is rarely the individual point defects induced by irradiation, instead it is the consequence of massive collective atomic displacements, such as segregation and diffusion (e.g. DeNatale and Howitt, 1984; Fan and Marks, 1989; Jiang and Silcox, 2002). Even if all the displaced atoms were radicals produced by the radiolytic processes, or affected by the radicals, the question remains as to how these species associate or dissociate collectively and rapidly. It appears that a type of swarm behavior causes the same species of atoms to move together coherently. In other words, there must be a mechanism other than the knock-on and radiolysis, and the operation of this mechanism is assisted by neither of the mechanisms.

In recent studies, strong evidence supporting the existence of electric fields induced by excitation and ionization processes in insulating materials has appeared (Jiang et al., 2002, 2003; Yamamoto et al., 2004 Gontard et al., 2012; Cazaux, 1995), and these electric fields are strong enough to displace atoms in a manner similar to that occurring in battery electrolytes (Jiang, 2015), affecting both long- and short-range order. The effects of the electric field have also been noticed previously in studies of hole-drilling (Humphreys et al., 1990) and migration of grain boundaries (Bouchet and Colliex, 2003) under a highly focused electron beam in scanning transmission electron microscopy (STEM). Accordingly, most of the damage phenomena caused by a STEM probe can be interpreted within the framework of damage by the induced electric field (DIEF) mechanism (Jiang, 2013).

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For STEM illumination, the lateral dimension of electron beam (e.g. <0.5 nm) is smaller than the effective mean-free-paths (MFPs) of SE and Auger electrons (e.g. >1.0 nm) (Seah and Dench, 1979), and thus it is reasonable to assume that the probed region is charged uniformly, along the beam direction, forming a positively charged nano-rod or nano-column (Jiang, 2013). Based on this assumption, the induced electric field for a given induced charge density ρ (Coulomb per length) can be simplified as (Jiang, 2013)

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

in which R is the shortest distance to the point of interest measured perpendicular to the beam. Thus the induced electric field has an approximately cylindrical symmetry around the beam and its strength is independent of specimen thickness (Jiang, 2013). This theory is supported by experimental observations of nano-cylinders (nano-rings in 2-D projection) in silicate glasses (Jiang et al., 2002, 2003; Gontard et al., 2012) and $Li_4Ti_5O_{12}$ (Su et al., 2013) formed by a STEM probe.

For transmission electron microscopy (TEM) illumination, the lateral dimension of the broad beam is usually larger than e.g. 10 nm, and thus it is comparable to or even much larger than the effective MFPs of emitted electrons. On average, the excited electron does not contribute to the charge accumulation if it is still in the illuminated region, unless it travels out of the region, by being emitted into vacuum from the surface. As a result, the unbalanced charges are no longer distributed uniformly inside specimen; instead, they are mainly trapped at the surface regions. So the induced electric field in TEM should be different from that in STEM, resulting in a different appearance of damage between TEM and STEM. Nevertheless, the DIEF mechanism has generally been overlooked in the broad beam illumination in TEM. In the recent review article (Jiang, 2016), it was extended to TEM illumination, and various damage phenomena observed in experiments can be interpreted by this mechanism.

In this paper, we derive the induced electric field for broad beam TEM illumination. Besides the current density of electron beam (Jiang, 2016), three other experimental parameters, which crucially affect beam damage in TEM by the DIEF mechanism, are identified as specimen thickness, beam size and wedge angle of a wedge-shaped specimen. Accordingly, the unique characteristics of beam damage in TEM are deduced, which include damage in the periphery of beam region, specimen thickness dependence, damage in the wedge-shaped specimen, edge smoothing effect, oxidation in non-oxide materials, and crystallographic orientation dependence. All these phenomena have been frequently observed in TEM studies, and can be readily interpreted in the framework of the DIEF mechanism.

2. Induced electric fields for collimated tem illumination

The calculation of the electric field in a specimen requires the charge density trapped at the surface region, which is unfortunately unknown. However, the distribution of electric field within the illuminated region can be evaluated qualitatively based on assumptions that the electron beam intensity is uniform (i.e. a top-hat approximation), so that the induced charges are trapped uniformly at the surfaces and thickness of the charged layers is ignored. These assumptions are in theory not necessary, but are only used for convenience to obtain analytical expressions for the electric field. Although the electric field under a non-uniform distribution will be different from the uniform assumption, it will show later that the qualitative conclusions may not be affected. Most importantly, the electric field model derived under these assumptions explains very well various damage phenomena observed in insulating materials in TEM.

Two types of specimen geometry are considered in this work: uniform thickness specimen and wedge-shaped specimen.

2.1. Homogeneous specimen with uniform thickness

The geometry of the illumination is illustrated in Fig. 1a. First, we assume that the electric field inside the specimen is induced by two equally charged parallel layers (or sheets), whose size is defined by the beam. (It should be pointed out that both surfaces may not be charged equally in reality, and this will be discussed later in this section.) For convenience of discussion, we assume that the charged region is a square with dimension of $2a \times 2a$, and the specimen thickness is $2T$. The beam direction is along to the $-\hat{z}$ direction. Here we only consider the electric field inside the specimen.

For a large (infinite) charged sheet, the electric field has uniform strength, $E_0 = \sigma/2\varepsilon_0\varepsilon_r$, in which σ is surface charge density (Coulomb per area) (Griffiths, 1999). Therefore, the electric field is zero in between two large (infinite) positively charged parallel sheets. This represents the extreme situation where $a \gg T$. In other words, the electric field insider a thin slab can be practically ignored at very low magnification in TEM. However, this simplification does not apply to the situation where the illuminated area is comparable with the specimen thickness, i.e. $a \sim T$. In addition, the induced electric field cannot be ignored near the edge of the electron beam.

Consider a surface element $dS = dxdy$on either the top or bottom surface, as shown in Fig. 1b. The charge carried by a surface element is $dQ = \sigma \times dxdy$. The electric field at a point $P(x_p, y_p, z_p)$ $(-T < z_p < T)$ inside the specimen produced by this element is

$$d\vec{\mathbf{E}} = \frac{\sigma}{4\pi\varepsilon_0\varepsilon_r} \times \frac{dxdy}{R^2} \times \hat{\mathbf{R}} = \frac{E_0}{2\pi} \times \frac{(x_p - x)\hat{\mathbf{x}} + (y_p - y)\hat{\mathbf{y}} + (z_p \pm T)\hat{\mathbf{z}}}{\left[(x_p - x)^2 + (y_p - y)^2 + (z_p \pm T)^2\right]^{3/2}} \times dxdy \tag{2}$$

in which $\pm$ represents element either at the bottom $(z_p + T)$ or the top $(z_p - T)$ surface. Here we use dimensionless variables, $x \to x/a$, $y \to y/a$and $z \to z/T$. The total electric field is a superposition of contributions from both surfaces, and can be calculated by integrating over two charged squares. Since σ is unknown, the evaluated electric field only gives its relative strength. For convenience, we set $E_0/2\pi = 1$. Then

$$E_x = \int_{-1}^{1} dy \int_{-1}^{1} dx \sum_{k=0,1} \frac{(x_p - x)}{\left[(x_p - x)^2 + (y_p - y)^2 + (T/a)^2\left(z_p + (-1)^k\right)^2\right]^{3/2}} \tag{3a}$$

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