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Intensity distribution analysis of cathodoluminescence using the energy loss distribution of electrons

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ABSTRACT

We present an intensity distribution analysis of cathodoluminescence (CL) excited with a focused electron beam in a luminescent thin film. The energy loss distribution is applied to the developed analysis method in order to determine the arrangement of the dipole locations along the path of the electron traveling in the film. Propagating light emitted from each dipole is analyzed with the finite-difference time-domain (FDTD) method. CL distribution near the film surface is evaluated as a nanometric light source. It is found that a light source with 30 nm widths is generated in the film by the focused electron beam. We also discuss the accuracy of the developed analysis method by comparison with experimental results. The analysis results are brought into good agreement with the experimental results by introducing the energy loss distribution.

1. Introduction

Cathodoluminescence (CL) excited by electron beam irradiation is used for many applications such as analysis of materials [1], field emission displays [2], laser sources [3] and light source of near field optical microscope and super resolution microscope [4–7]. Cathodoluminescence analysis is a crucial technique for understanding the optical properties of nanowires [8–10], nanodiamonds [11–14], nanoparticles [15,16], and plasmonic nanoantennas [17,18] because it has nanometric spatial resolution. The spatial resolution of CL analysis is much higher than that of photoluminescence techniques because the electron beam can be focused to regions a few nanometer in size. The spatial resolution of CL analysis is determined by the spatial extent of electron beam scattering and light propagation excited with the electron beam. In order to evaluate and improve the spatial resolution of CL analysis, it is required to analyze the scattering of electron beam and the light propagation of CL.

We have developed a new analysis method for the light intensity distribution of CL using a combination of Monte Carlo simulation and the finite-difference time-domain (FDTD) method [19]. The analysis method has three steps. The first step is a scattering analysis of the incident electron beam by Monte Carlo simulation [20–22]. The trajectory and the energy loss of each electron are simulated with the analysis method. The second step is the calculation of the arrangements of dipole excited with the electron beam. The position of the dipole is determined by the trajectory of incident electron analyzed by Monte Carlo simulation. CL emission is expressed as dipole radiation in the analysis method. Finally, light propagation from each dipole is analyzed by the FDTD method. The intensity distribution of light emitted from each dipole is summed up to obtain the intensity distribution in and near the thin film.

In this study, we propose a new analysis method for the intensity distribution of CL in a luminescent thin film that incorporates the energy loss distribution of the electrons [23,24]. This analysis method is based on the Monte Carlo simulation and the FDTD method. The dipole locations are placed at the multiple positions along the trajectory of the electron. We analyze the intensity distribution of CL in the luminescent film and evaluate the accuracy of the analysis through a comparing with experimental results.

2. Material and methods

2.1. Principle of the electron beam scattering analysis

Fig. 1(a) shows the principle for electron-beam scattering analysis by Monte Carlo simulation. When the electrons irradiate a luminescent film, the electrons interact with the electrical fields of

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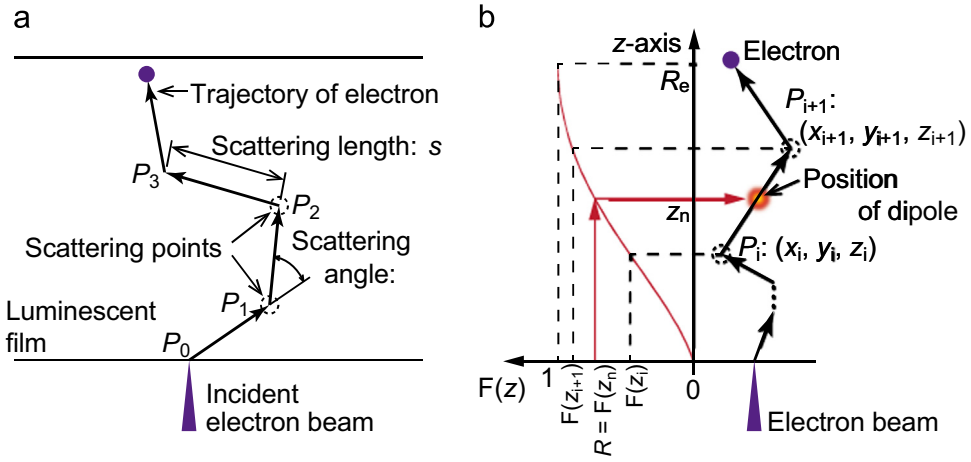

Fig. 1. (a) Schematic of the principle of electron beam scattering analysis by Monte Carlo simulation. The length that the electron travels between scattering events and scattering angle are calculated in a Monte Carlo simulation. (b) Schematic of the positioning of dipole based on the energy loss distribution of electrons. The dipoles are positioned between scattering points P_i and P_{i+1}. The position of each dipole is determined by using a random number R between 0 and 1.

the atoms and are scattered by the atoms.

In the Monte Carlo simulation, the electron trajectories are determined as follows. As shown in Fig. 1(a), the electron is initially incident upon P_0 on the bottom surface of the substrate. The electron is scattered at P_0 and travels to P_1; the electron is scattered again at P_1 and repeats this process until the electron loses all of its energy. The electron is assumed to scatter elastically. The scattering angle ϕ is determined by

$$\cos\phi = 1 - \frac{2\alpha R}{(1+\alpha-R)}$$

where R is a random value and α is the scattering factor. The random number is constrained to be between 0 and 1. The scattering factor α is given by

$$\alpha = 3.4\times10^{-3}\frac{Z^{0.67}}{E}$$

where E is the energy of the electron, Z is the atomic number of the luminescent thin film material. The distance traveled between sequential scattering positions s is determined by

$$s = -\lambda \log_e R$$

The λ is the mean free path of the electron. The λ is given by

$$\lambda = \frac{A}{N_a\rho\sigma_E}$$

where N_a is Avogadro's number, ρ is the density, and A is the atomic weight of the luminescent thin film. σ_E represents the scattering cross-section and is given by

$$\sigma_E = 5.21\times10^{-21}\frac{Z^2}{E^2}\frac{4\pi}{\alpha(1+\alpha)}\left(\frac{E+511}{E+1024}\right)^2$$

The electrons lose their energy when they scatter in the luminescent film. The energy loss is due to electrostatic force and inelastic scattering. The energy loss rate of the electron, dE/ds, is given by [25]

$$\frac{dE}{ds} = -78500\times\frac{Z\rho}{AE}\times\log_e\left(\frac{1.166(E+0.85J)}{J}\right)$$

where J is the mean ionization potential, which is calculated from

$$J = \left(9.76Z + \frac{58.5}{Z^{0.19}}\right)\times10^{-3}$$

The calculation of the distances traveled and the scattering angles are repeated until the energy of the electron is sufficiently low. The numbers of generated dipoles between sequential two scattering points, N, is given by [26]

$$N = \frac{\Delta E}{3E_{BG}}$$

where ΔE is the energy loss of the electron and E_{BG} is the band gap energy of the luminescent thin film. We considered the dipoles to be not excited if N was less than 1.

2.2. Arrangement method of generated dipoles

The generated dipoles are arranged along the trajectory of the electron. In the previous analysis method reported in Ref. [19], the generated dipoles are positioned at the midpoints between P_i and P_{i+1}. The analysis gives good results for relatively thicker films and low acceleration voltages. However, the technique is unable to analyze the case of high acceleration voltages and very thin films because the distance that the electron travels becomes relatively longer than the film thickness and many dipoles tend to be placed at middle of the thin film. Modification of the dipole arrangement method is required to improve the accuracy of the analysis.

In the new method, the generated dipoles are positioned at the multiple locations between P_i and P_{i+1}. To calculate the dipole positions, we use the energy loss distribution of electrons [23], which is given by

$$g(L) = 0.6 + 6.21L - 12.4L^2 + 5.69L^3$$

where L is the normalized penetration depth of the electron. L is given by z/R_e where z is the distance from the film surface. R_e is the maximum range of the electron penetration [27], and calculated according to

$$R_e = (27.6A/\rho Z^{0.889})E^{1.67}$$

Fig. 1(b) shows the principle used to arrange multiple dipoles along an electron trajectory. The function $F(z)$ represents the probability distribution function of generated electrons along the z-axis and is calculated as

$$F(z) = \frac{\int_0^{z/R_e} g(L)dL}{\int_0^1 g(L)dL} \quad (1)$$

$F(z)$ varies from 0 to 1 because z in Eq. (1) can vary between 0 and R_e. As shown in Fig. 1(b), we suppose one electron is scattered at