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SIMULATION OF SECONDARY ELECTRON TRANSPORT IN THIN METAL AND FULLERITE FILMS

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Excitation processes and transport of recoiled and secondary electrons generated in fullerite and metal films under photons and electron irradiation were studied by computer simulation. Studied processes resulting in polymerization of fullerite were considered as the basic ones in formation of a pixel in electron nanolithography with fullerite film as an electron-beam resist. Reliability of the computer model and the important role of secondary electrons in the process of pixel formation were confirmed by comparison of the sizes of the calculated secondary electron swarm and the experimental cluster-pixel obtained previously. The photoelectron yield dependence on the incident photon's energy was also obtained with the same computer model for metal foils which can be used as a radiation strip-detector.

Keywords: Fullerite, Films, Electron beams, Electronic properties.

1. Introduction

Studying primary and secondary electron transport in metal films and in fullerites is important both for improving the efficiency of strip detectors [1] and for better understanding the mechanisms of fullerite modification and pixel formation by electron nanoprobe in high resolution electron beam lithography with fullerite as an electron beam resist [2–6]. Computer simulation of the transport for all electrons generated in fullerite C_{60} under irradiation of primary electrons in the keV-energy range has been developed in the framework of the Monte-Carlo model [4, 5]. A possible important role for secondary electrons in the formation of lithographic images is assumed to be in the process of formation of the minimal image in the form of a point (pixel) by the electron nanoprobe in a fullerite C_{60} film [5]. The developed computer model was also applied to describe the charge accumulation in metal strip detectors [1]. In this paper, the developed computer model was applied in two cases: first, to obtain information about photoelectron yield and charge accumulation needed for strip detector design and second, to clarify the role of secondary electrons in pixel formation in a fullerite C_{60} film by comparing the model results with experimental data.

2. Model and experimental details

According to [4, 5], the computer model included the Monte-Carlo method for both electron transport description using the cross-sections for electron-atom elastic collisions with carbon (or nickel) atoms as well as for impact ionization. To describe photo-ionization, we have translated Penn's model [7] for electrons with full energy loss and have taken into account surface plasmons. All simulations were managed by the developed program packages. The program works as follows: electrons generated by ionization of atoms, in turn, trigger a number of collisions, which can also lead to the emergence of the next generation of electrons, etc. Once the particle has passed the distance l, distributed by the law $e^{-l/\lambda}/\lambda$, where λ is the mean free path length, it dissipates via one of the interaction processes. The probability of the i-th process by which scattering a particle in a given state is equal σ_i/σ_{tot}, where σ_i is the corresponding cross section and σ_{tot} is the sum of those cross sections. In the evolution cascades, a crucial role is played by the collision's probability, thus only the largest cross sections in the above energy range were considered.

Modeling the irradiation processes by photons with 0.1–15 keV energy showed that photons also generate a swarm of secondary electrons. The number of these electrons is proportional to the number of photons and their energy satisfies the photoeffect law. The probability of photoelectron generation decreases exponentially in the target depth, and their further transport is determined only by interaction with the target. The principal distinction between photon and electron collisions with the targets atoms was well traced in the simulation. One photon is absorbed only once, creating a high-energy electron, and then it no longer interacts with the target. An electron with the same energy undergoes a huge number of collisions with the target atoms, and can transfer energy in a wide range, hence it can modify the target properties in the large volume. Numerous inelastic collisions between secondary electrons and carbon atoms in fullerite lead to creation of chemical bonds between fullerenes and finally to fullerite polymerization [5]. Secondary electrons near the foil surface can escape and thereby enhance the sensitivity of a metal-strip detector.

Modeling the electron transport in fullerite C_{60} has been performed using previously described experimental conditions [6] to compare the calculated and experimental data. A film with a thickness of 200–400 nm was grown by evaporation of high-purity soot onto the Si-substrates. Then, the film was irradiated by an electron nanoprobe with diameter less than 10 nm in a set of points to create a hidden image in the form of point clusters due to polymerization of irradiated areas. The energy of the irradiating electrons was 15 keV. Then, the irradiated film with a latent image was developed in chloroform. Irradiation and visualization of samples after development were made by a Scanning Electron Microscope [6].

Modeling the photo-irradiation processes was oriented to the experiments performed with a new kind of X-ray sensor that can potentially be used as an X-ray beam position monitor. It has been developed at the Kiev Institute for Nuclear Research and has been tested on the B16 beamline [8]. The sensitive part of this device is a nickel foil divided into four quadrants which are independently measured using four channels of a 4-channel low current monitor. Measurements have been performed to characterize the performance of the device as an X-ray Beam Position Monitor (XBPM) by reading the photocurrent emitted from the foils.

3. Results and discussion

The computer model was first applied to the relatively simple problem of photoionization yield description. Fig. 1 demonstrates the energy dependence of photoionization yield for a 50 μm nickel foil obtained in [8] (dashed curve) and in our simulation (solid curve). The low yield scale (of about 10^{-3}) is a result of high target density which reduces the secondary electron mean free path. Therefore, the calculated maximal radial deviation of secondary electrons was as low as 60–80 nm, which restricts the number of electrons leaving the nickel film. The minimum in the $h\nu = 8 - 10$ keV photon energy range is conditioned by the 8.3 keV nickel atomic level. Its excitation results in essential photon loss. Therefore, the number of electron-hole pairs, generated mainly by photons in conductivity zone, decreases along with the photoionization yield. Comparison of simulation and experimental data [8] shows satisfactory agreement of calculated and experimental data and the validity of the proposed model.

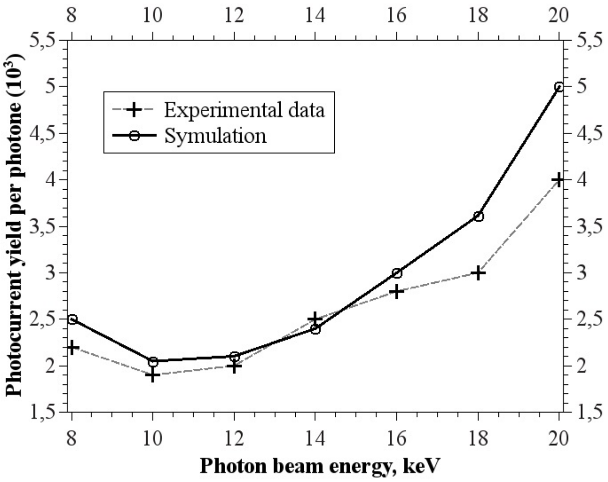

FIG. 1. Photoionization yield of nickel foil of 50 μm thickness obtained in experiment [8] (dashed curve), and in simulation (solid curve)

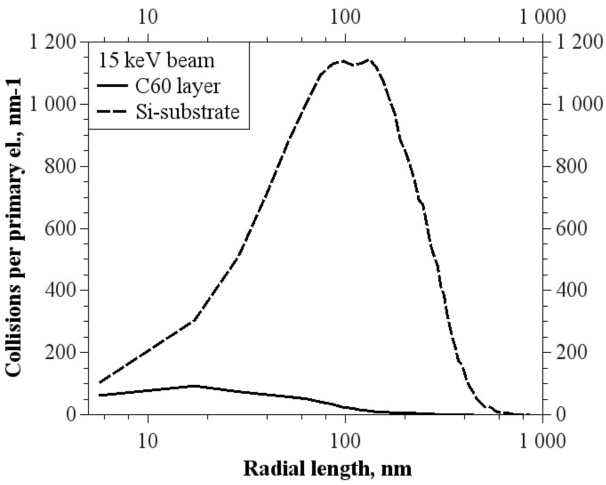

FIG. 2. Radial distributions of the inelastic collisions density between primary 15 keV electrons and atoms in 300 nm layer fullerite and in the Si-substrate

The fullerite film used in the experiment [6] was not thick enough to absorb irradiating (primary) 15 keV electrons. Therefore, the majority of primary electron-atom collisions followed by secondary electron generation and collisions takes place in the Si-substrate, which is confirmed by Fig. 2. This figure shows the calculated radial distribution of the density of all inelastic electron-atom collisions induced by 15 keV primary electrons in fullerite film and Si-substrate. We assume that every inelastic electron collision which transfers more than 2 eV to the fullerene electron system can excite it, and therefore, with some probability, create a chemical bond between two neighboring fullerenes. Therefore, the inelastic collision density can be considered a measure of the polymerization rate. In inelastic collisions, electrons with sufficient energy (more than about 8 eV) can also generate new secondary electrons. The energies of the majority of secondary electrons do not exceed several eV. Therefore, their inelastic cross section is very small and lifetime is very large. By this reasoning, the low-energy secondary electrons penetrate from silicon substrate back into the fullerite film. One should consider the substrate as an intensive source of low-energy electrons which can essentially enhance the polymerization of a fullerite film and enlarge the size of lithographic pixel in it. Note, that the radial distribution of inelastic secondary electron-atom collisions in the above-mentioned nickel foil, induced by the 15 keV photons, is similar to the one shown in Fig. 2, but the number of collisions is nearly three orders of magnitude less .

Figure 3 shows the radial distribution of the fullerite film-based inelastic electron-atom collision density for two groups of electrons: the low-energy group (2–10 eV) and the high-energy group (10 eV -E), where the energy of primary electrons E was 15 keV and 10 keV, respectively. One can see that the collision density for low-energy electrons does not radically exceed that for high energy ones near the beam axes (in several times only). But away from the primary beam area, collisions associated with low-energy electrons qualitatively dominate by many orders of magnitude, due to their huge numbers. Modeling showed that the effect of secondary electron generation by the substrate is smaller for the 10 keV beam, and it diminishes in energy due to a shortening of the penetration depth of the primary electrons. At the low beam's energy, the secondary electrons are generated near the surface and can thus leave it immediately.

The role of secondary electrons and the substrate is more important in the case of photo-irradiation. The fullerite film is considered to be transparent for 15 keV photons. Therefore, practically all secondary electrons are formed in deeper layers of the Si-substrate. However, as is shown by our calculations, the electrons penetrate into fullerite film from the substrate. In addition, the collision density induced by photons is much smaller in fullerite film because secondary electrons are generated in deeper substrate layers.

The results of computer simulation for electron transport under irradiation of a 15 keV electron nanoprobe represented in Fig. 2 and 3 allow qualitative explanation of the corresponding experiment [6] – see Fig. 4. In this experiment, compound carbon clusters, consisting of a large disk-shaped cluster with small point-cluster superimposed on the top in the center, were formed.

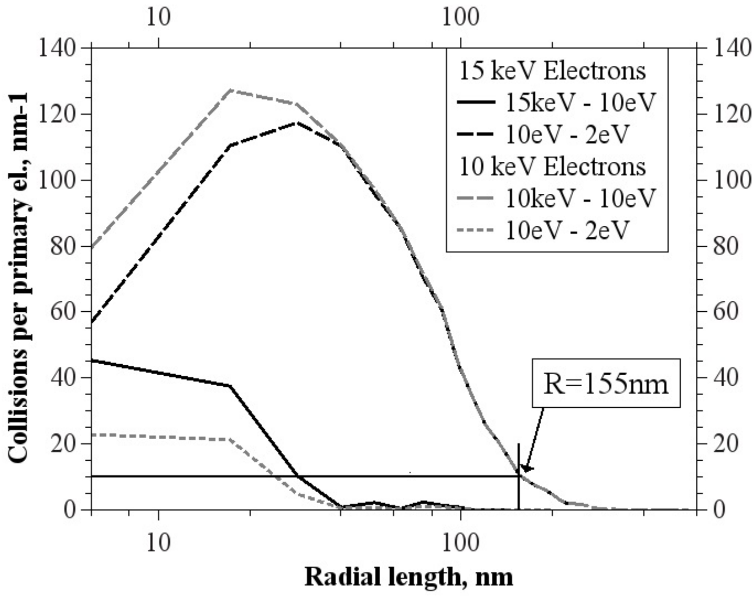

FIG. 3. Radial distributions of the inelastic collisions of all electrons with energy $>$ 10 eV and energy $<$ 10 eV with atoms. Primary electron energy is 10 and 15 keV

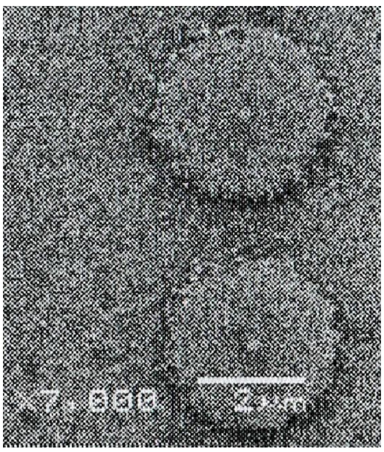

FIG. 4. Carbon clusters formed in the 300 nm fullerite film on the Si-substrate by a nanoprobe with energy 15 keV [6]. Cluster radius is about 1.5 microns

The simulation data qualitatively confirms the assumption that small clusters in the center are formed mainly by the high-energy electrons, while the large disk cluster is originated due to the polymerization under a swarm of low-energy secondary electrons. However, the radius of the disk-shaped cluster (~ 1500 nm) is much larger when compared to the secondary electron radial distribution obtained in the simulation ($\sim 350 - 400$ nm). While this result may seem incongruous, there is an explanation. One more effect should be taken into consideration to explain the larger size for the experimental pixel. It is a modification of fullerite properties (polymerization) during irradiation. Creation of new chemical bonds in a completely polymerized area is impossible. This fact can be considered by gradually reducing the inelastic electron collisions cross section and by correspondingly increasing the electron mean free-path during irradiation. Then, more of the secondary electrons from the cluster area may be transported to the cluster periphery, resulting in a radical increase of the polymerization rate there and thus enlargement of the cluster radius. This effect has been simulated and it was determined that the maximal and average spreads of low-energy electrons increase in dependence on the reduction of the relative inelastic cross section. It was shown that electron spread can reach the experimental radius value $R = 1500$ nm at one third of the initial cross section which corresponds to the creation of a majority part of the possible polymeric bonds.

4. Conclusions

Simulation using the developed computer model confirms the assumption [3, 6] that low-energy secondary electrons play an important role in fullerite polymerization and pixel formation in electron lithography with fullerite as an electron beam resist. The total contribution of secondary electrons to this modification can exceed 90%, and their radial deviation (the pixel radius) can be one to two orders of magnitude larger than the radius of the primary electron beam. A new mechanism for the radical increase of the pixel radius is suggested. This implies a long distance spread of the low-energy secondary electrons with reduced energy losses due to gradual polymerization of the irradiated area. The photoionization yield of metal foils is also caused due to secondary electron transport. A dependence of the nonmonotonous yield upon photon energy was observed and properly described by the modeling.

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