Modeling of Multi-Time Scale Particles in Rarefied Gas and Plasma

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Abstract. Self-sputtering is a promising technology for a forthcoming copper wiring of semiconductors. In the discharge space of self-sputtering apparatuses, there exist electrons, copper ions, and copper atoms. Since the velocities of three kinds of species are different in order, they are multi-time scale particles. The difficulty in the analysis of the self-sputtering is in the strong coupling of motion between fast and slow particles.

The self-sputtering of copper was simulated by a simultaneous modeling of electrons, ions, and atoms. It was found that the number density of copper atoms is largely nonuniform. Also it was found that reflection of parts of copper atoms and ions on the electrode is essential in sustaining the discharge in the self-sputtering.

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INTRODUCTION

Wiring of semiconductors is changing from aluminum to copper because the communication speed for copper wiring is higher than that for aluminum. Efficient copper wiring is done best by using the self-sputtering. That is, copper atoms that are sputtered from the copper target are used as a discharge gas. This new technology has yielded a new academic problem. In conventional sputtering using argon gas, gas pressure is about 0.7Pa and hence, the number density of argon atom is much higher than that of electron or ion. This means that the flow field of argon atoms, which are slow particles, is almost decoupled with the filed of plasma consisting of fast particles. The situation makes it possible to analyze plasma separately from gas flow. This is not true in the self-sputtering. The discharge gas consists of the copper atoms that are generated by the sputtering of copper target by the bombardment of higher energy copper ions. That is, the state of bombarding ions governs the density field of slow copper atoms, and moreover, the latter governs the production rate of plasma because ionization rate is proportional to the density of copper atoms.

In this work we study the copper self-sputtering by examining the motions of electrons, ions, and atoms simultaneously. Although the velocities and hence, time-scale of three species are different in order, the simultaneous simulation is indispensable. We use the particle modeling based on, so to speak, PIC/MC + DSMC[1].

1-D MODEL OF SELF-SPUTTERING

Particle simulation of multi-time scale particles is computationally intensive. We start from one-dimensional problem. That is, the electric field is one-dimensional. Let $\phi(z)$ be the potential. The copper target is at $z = 0$ and the substrate is at $z = L$, where L is the electrode distance. For simplicity it is assumed that a uniform magnetic field B is applied in the x-direction. The target is powered by a radio frequency voltage and the substrate is grounded. The species in the discharge space are electron (e^-), ion (Cu^+), and atom (Cu).

The phenomena in the space are

(a) motion of particles in electric and magnetic fields,
(b) e^--Cu, Cu^+-Cu, and Cu-Cu collisions, disregarding Coulomb collisions,
(c) sputtering of the target by Cu^+ and Cu,
(d) reflection of Cu^+ and Cu on the electrodes,
(e) secondary electron emission by the bombardment of Cu^+,

(f) change in electric field due to the change in charge density.

MOTION OF PARTICLES

We want to determine the state at time $t+\Delta t$ by using known properties at time t. The equation of motion is

$$m\frac{d\vec{v}}{dt} = q(\vec{E}+\vec{v}\times\vec{E}) \equiv \vec{F} \tag{1}$$

where m is the mass of a particle with charge q, $\vec{v}$ is the velocity, $\vec{E}=(0,0,E)$ is the electric field, and $\vec{B}=(B,0,0)$ is the magnetic field. In case of rf discharge, E depends on the field variable z and time t, i.e., $E(z,t)$. Equation (1) can be stably solved by use of the modified Verlet method. The z-coordinate of a particle is

$$z(t+\Delta t) = z(t) + v_z(t)\Delta t + \frac{(\Delta t)^2}{2m}F_z(t), \tag{2}$$

where $F_z(t) = q(E(t) - v_y(t)B)$ and $E(t)$ denotes $E(z(t),t)$. The new positions for all charged particles (e^- and Cu^+) yield the field of charge density $\rho(z,t+\Delta t)$, and hence, we have the field $\phi(z,t+\Delta t)$ from the finite difference form of the Poisson equation. Then we have

$$\vec{F}(t+\Delta t) = q(\vec{E}(t+\Delta t) + \vec{v}(t+\Delta t)\times\vec{B}). \tag{3}$$

Substituting eq.(3) into the form

$$\vec{v}(t+\Delta t) = \vec{v}(t) + \frac{\Delta t}{2m}[\vec{F}(t)+\vec{F}(t+\Delta t)], \tag{4}$$

we have $\vec{v}(t+\Delta t)$. In PIC/MC simulation the procedure to obtain $\rho(z,t+\Delta t)$ is called the charge assignment.

COLLISIONS BETWEEN PARTICLES

Among 3 types of Coulomb collisions, $e^- - e^-$ collisions have a dominant effect on the electron energy distribution function in plasmas[1]. The effect becomes larger as the electron density increases. It is shown that the effect is very small at the electron density of $10^{17}m^{-3}$[2]. This density is smaller the density considered in this paper. Therefore we disregard the Coulomb collisions.

It is probable that some particles collide with others in $(t,t+\Delta t)$. Using the velocities $\vec{v}(t+\Delta t)$ at the end of motion, we consider the collisions in $(t,t+\Delta t)$. This procedure requires that the collision probability in Δt is much smaller than unity. Since e^--Cu, Cu^+-Cu, and Cu-Cu collisions are of short range ones, we use the maximum collision number method[1]. Cu and Cu^+ are treated as rigid sphere. Let (i,j) be a collision pair, where i and j belong to unlike species. We first divide the computational domain $(0 \le z \le L)$ into small cells with volume V_c. Let N_i and N_j be the numbers of simulated particles of species i and j, respectively. The maximum number of collisions between species i and j in a cell in time Δt is

$$N_{\max} = \max(W_i,W_j)N_iN_jV_c^{-1}(g_{ij}\sigma_{ij})_{\max}\Delta t, \tag{5}$$

where W_i is the weight of particle i, g_{ij} is the relative velocity between particle i and j, and σ_{ij} is the total collision cross section. For collisions between like species i, we have only to replace N_j by $(N_i-1)/2$ and $\max(W_i,W_j)$ by W_i.

Cu-Cu collisions

Let σ_{ij} be πd_i^2, where d_i is the diameter of Cu atom. For each cell we consider $N_{\max}$ tentative collisions between randomly chosen pairs. Let g_{ij} be the relative velocity of such a pair. The pair is let to collide with probability $g_{ij}/g_{\max}$. If collision occurs, it is isotropic.

e^--Cu collisions

Since the velocity of e^- (species i) is much higher than that of Cu (species j), eq.(5) takes the form

$$N_{\max} = \max(W_i, W_j) N_i N_j V_c^{-1} (v_i \sigma_{ij})_{\max} \Delta t, \tag{6}$$

where v_i is the speed of e^- and σ_{ij} depends only on the kinetic energy ε of e^-. We write $v\sigma_T$ in place of $v_i\sigma_{ij}$. The total cross section σ_T is the sum of the cross sections for elastic collision, exciting collision, and ionizing collision, shown in Fig.1[3]. The value of $(v\sigma_T)_{\max}$ is evaluated by use of Fig.1. We sample $N_{\max}$ electrons randomly from a cell. Repeat

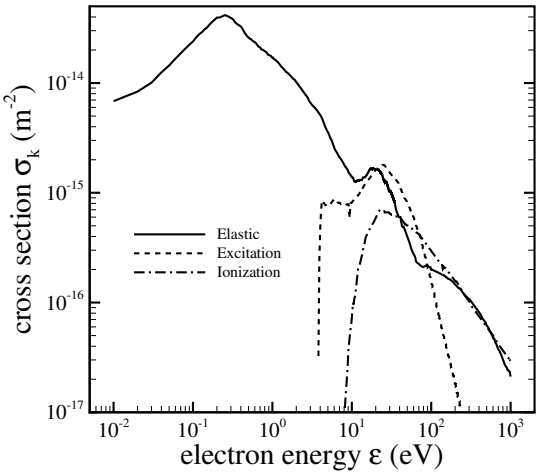


FIGURE 1. Cross-section for electron - Cu collision

the following judgment for each electron. Let v_i be the velocity of a sampled electron. If both $U < v_i\sigma_T/(v\sigma_T)_{\max}$ and $U' < W_j/\max(W_i, W_j)$ are satisfied, the electron is let to collide with Cu atom, where U and U' are independent uniform random numbers between 0 and 1. If a collision occurs, the type of collisions is determined in proportion to three cross sections in Fig.1.

Cu^+-Cu collisions

First we randomly choose $N_{\max}$ pairs (Cu^+, Cu) in a cell. Then we repeat the collision judgment for each pair as follows. Let g_{ij} be the relative velocity of pair (i, j), where i denotes Cu^+ and j denotes Cu. If $U < g_{ij}/g_{\max}$, the pair has a possibility of collision. The true collision occurs with probability $W_j/\max(W_i, W_j)$ for particle i and with probability $W_i/\max(W_i, W_j)$ for particle j. If a true collision occurs for particle i, it is the elastic collision or charge-exchange collision [4] with equal probability.

BOUNDARY CONDITIONS FOR PARTICLES

In a conventional sputtering, discharge is sustained by the ionization of argon atoms. In the self-sputtering, the way how incident Cu^+ and Cu behave on the surface of the electrodes plays an essential rule in sustaining discharge. We found from preliminary calculations that if all Cu^+ and Cu that are incident on the two electrodes are stuck to the surface, the discharge is impossible. This led us to the conclusion that incident Cu^+ and Cu must recoil with some probability. Of course, when Cu^+ recoils, it becomes Cu. Copper atoms are also supplied into the discharge space by the sputtering of copper target. The loss mechanism of Cu atoms is sticking onto the electrodes and ionization due to e^--Cu collision. Balance of supply and loss of Cu atoms results in a steady state.

Let W_e, W_i, and W_a be the weight of simulated particles e^-, Cu^+, and Cu. When Cu^+ is incident on the target, a secondary electron is emitted with probability $\gamma(W_i/W_e)$ and a sputtered atom is emitted with probability $Y(W_i/W_a)$,

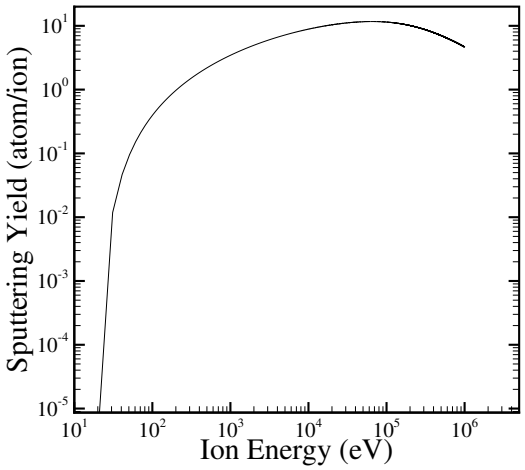


FIGURE 2. Sputtering yield Y (atoms/ion)

where γ is the secondary electron emission coefficient and Y in Fig.2 is the sputtering yield [5]. The incident ion itself recoils with probability $P_{\rm rc}$ or is stuck to the surface with probability $(1-P_{\rm rc})$. In case of recoil, a new Cu appears with probability $P_{\rm rc}(W_{\rm i}/W_{\rm a})$. When a Cu atom is incident on the target, the atom recoils with probability $P_{\rm rc}$ and a sputtered atom is emitted with probability Y. For the grounded electrode, we set $\gamma = Y = 0$ and assumed the same $P_{\rm rc}$ for Cu and Cu^+.

COMPUTATIONAL CONDITION

The radio frequency f is set 13.56 MHz, the voltage amplitude $V_{\rm rf}$ is 1000V, the magnetic field B is 0.01T, the electrode distance L is 50mm, and the secondary electron emission coefficient γ is 0.1. Introduction of magnetic field made it easier to sustain the discharge. Although initial conditions have no effect on final results, appropriate choice reduces computational task. Our choice is as follows : Cu pressure is 0.05Pa, e^- density and Cu^+ density are $10^{-16}{\rm m}^{-3}$. Initially Cu, e^-, and Cu^+ are assumed to be in equilibrium.

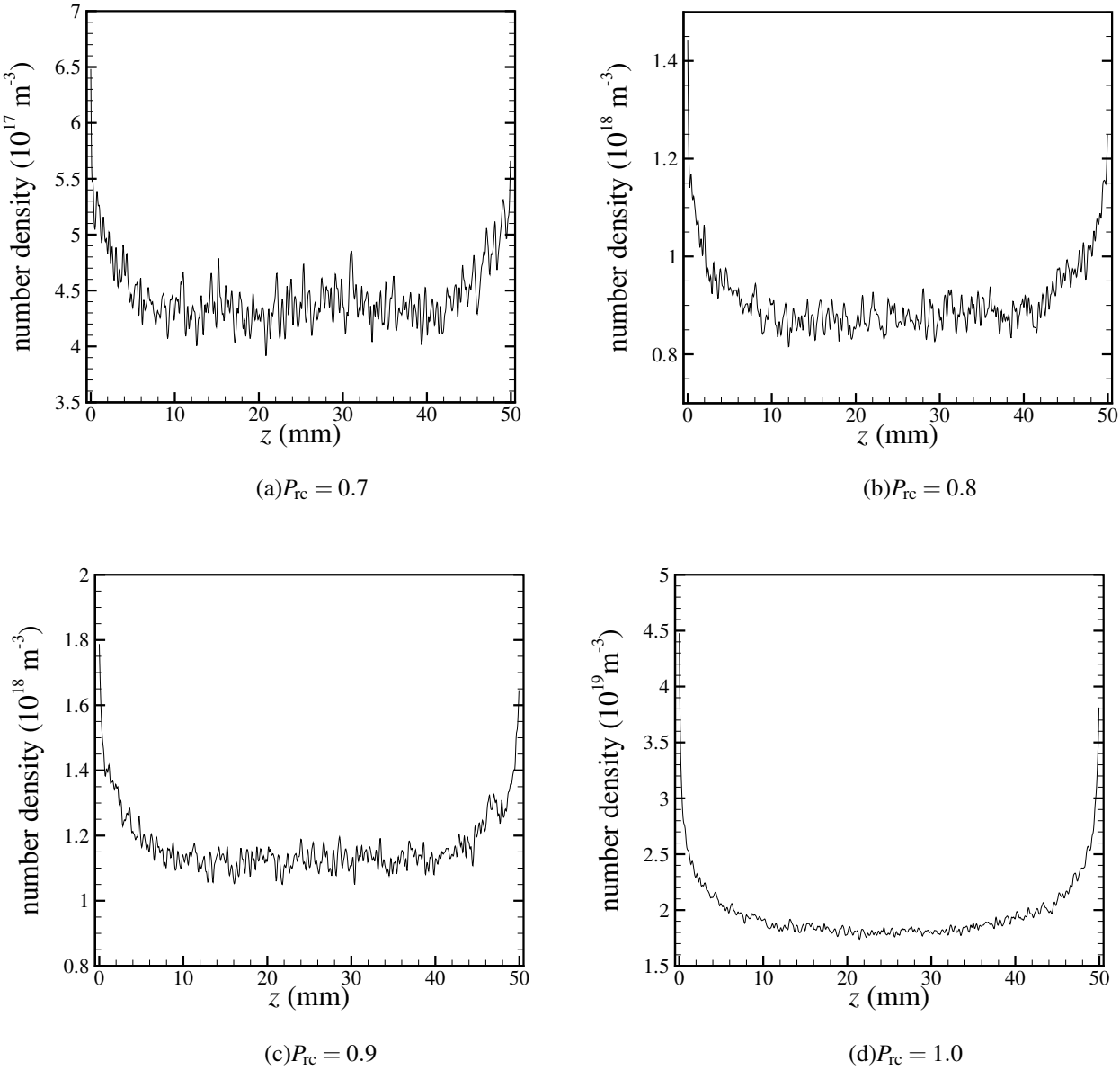
The timestep of electron is chosen as $\Delta t_{\rm e} = f^{-1}/960$. For this $\Delta t_{\rm e}$, the collision probability of electron is at most 0.1. The timestep of ion is chosen as $\Delta t_{\rm i} = 10\Delta t_{\rm e}$. The timestep of atom is set equal to $\Delta t_{\rm i}$. The number of grids is 1000, i.e., $\Delta z = 0.05$mm. In a final stage of the time development, the discharge goes into a periodic steady state. The time-averaged data are sampled by fixed phase ωt.

STRUCTURE OF SELF-SPUTTERING

The main concern is the effect of recoil probability $P_{\rm rc}$ on the structure of self-sputtering. We considered four cases of $P_{\rm rc} = 0.7, 0.8, 0.9$,and 1.0. Figure 3 shows the number density $n_{\rm a}$ of Cu. The overall level of $n_{\rm a}$ increases with $P_{\rm rc}$. Clearly, this is due to less sticking for larger $P_{\rm rc}$. Also the equation of state gives that pressure of Cu is very low, e.g., 0.002Pa for $P_{\rm rc} = 0.7$ and 0.08Pa for $P_{\rm rc} = 1.0$. This is much lower than 0.6Pa of argon gas in conventional sputtering. Measuring the partial pressure of metal vapor such as Cu is very difficult. We were able to predict Cu pressure by our modeling. Also, it was found that the density field of Cu is strongly nonuniform. The density is highest near the target because Cu atoms are supplied by the sputtering in addition to the recoil of incident Cu^+ and Cu.

Figure 4 shows the time-averaged electron density $n_{\rm e}$. The ion density $n_{\rm i}$ is equal to $n_{\rm e}$ except in the sheaths. In case of $P_{\rm rc} = 0.7$, the density $n_{\rm e}$ in the plasma bulk is about 10% of atom density $n_{\rm a}$. This means that the state of atom is strongly coupled with the state of electrons. We see that the probability $P_{\rm rc}$, by which Cu density ($n_{\rm a}$) is strongly influenced, has little effects on the electron density. In case of $P_{\rm rc} = 1$, the profile of $n_{\rm e}$ is somewhat higher and wider than other cases. This is consistent with Fig.3, where $n_{\rm a}$ for $P_{\rm rc} = 1$ is much higher than other cases.

Figure 5 shows the electric field E for various phases ωt of one radio-frequency cycle. Note the voltage of the target at $z = 0$ is given by $1000\cos\omega t$. The dependence of $E(z, \omega t)$ on z is quite different from that for one-dimensional rf discharges of argon [6]. Excepting the case of $P_{\rm rc} = 1$, the field E at $\omega t = 0$ and π is nearly symmetrical with respect to the mid plane $z = L/2$. In case of $P_{\rm rc} = 1$, the profile of E somewhat approaches to that for the argon discharge.



(a)$P_{\rm rc} = 0.7$ (b)$P_{\rm rc} = 0.8$

(c)$P_{\rm rc} = 0.9$ (d)$P_{\rm rc} = 1.0$

FIGURE 3. Number density of Cu atom (m^{-3})

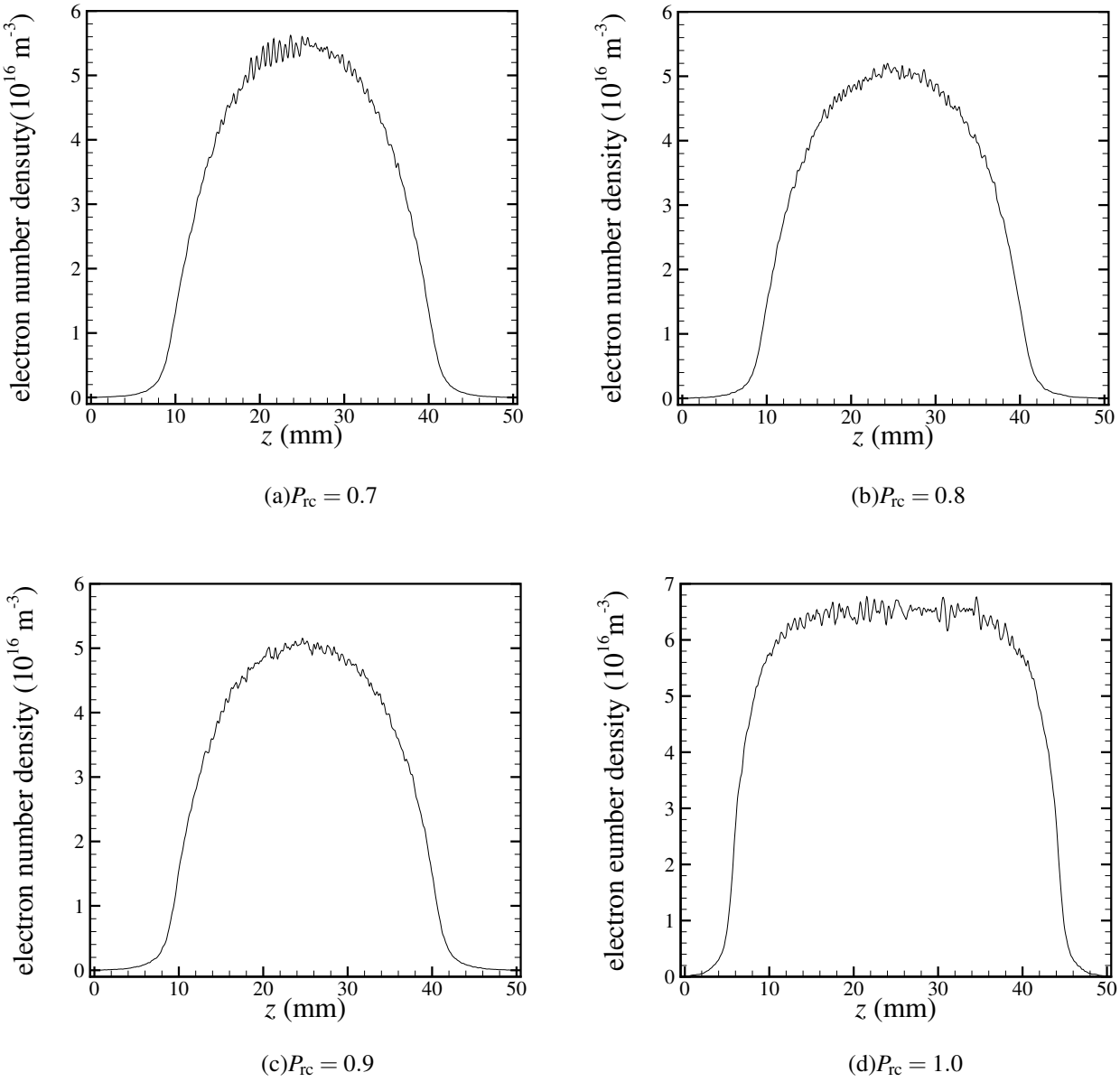
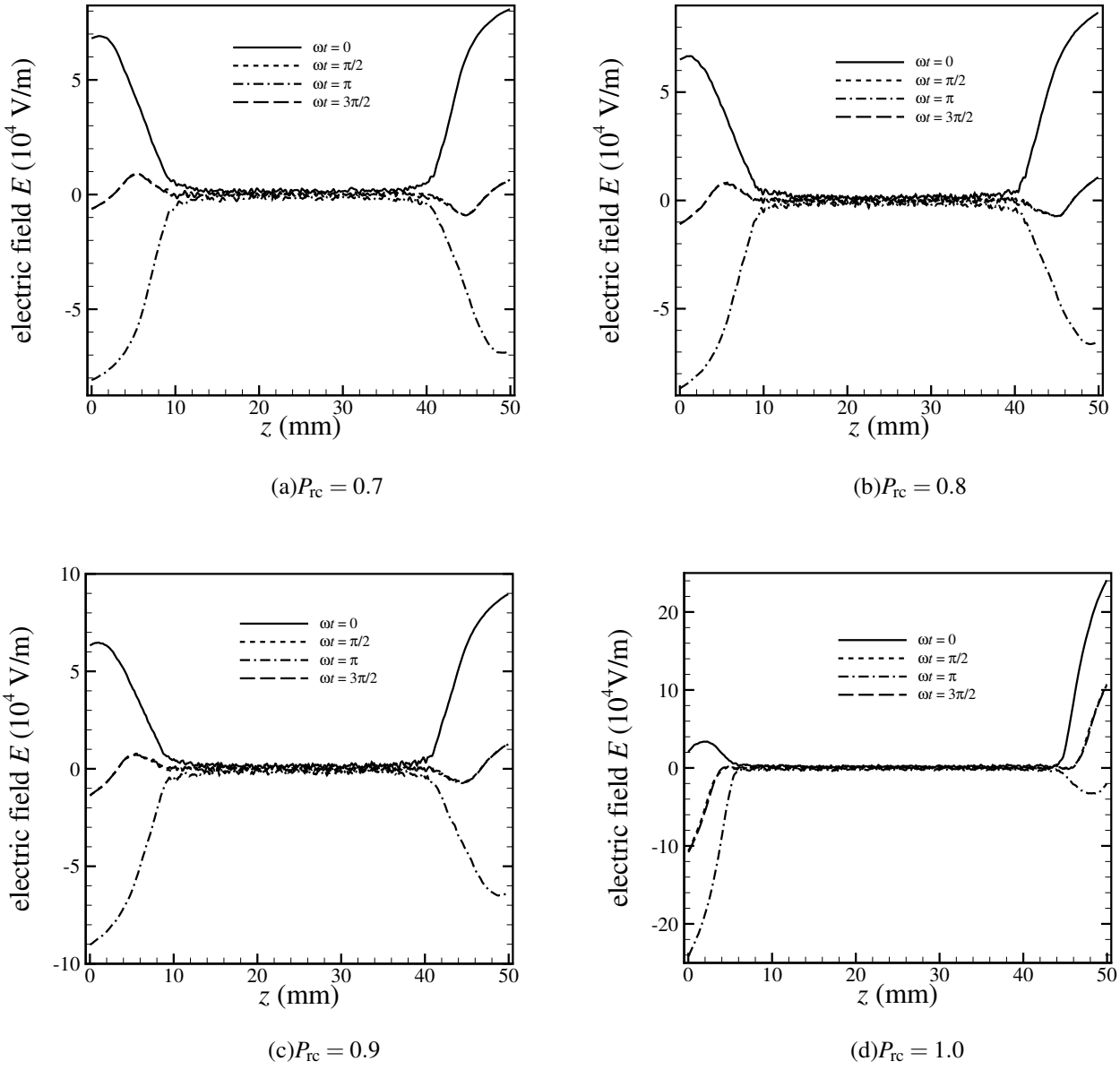


FIGURE 4. Number density of electron (m^{-3})



(a)$P_{\rm rc} = 0.7$ (b)$P_{\rm rc} = 0.8$

(c)$P_{\rm rc} = 0.9$ (d)$P_{\rm rc} = 1.0$

FIGURE 5. Electric Field E (V/m)

CONCLUDING REMARKS

In the self-sputtering for copper wiring, plasma consists of disparate velocity particles such as electrons, ions, and neutral atoms. Owing to the disparate velocities among species, the self-sputtering becomes one of the multi-time scale phenomena, which are difficult to solve. We clarified the structure of the self-sputtering by intensive use of computer to give a touchstone for future studies of others. It is expected that some elegant ideas are introduced in a near future.

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