PIC/MCC analysis of a photoresonance plasma sustained in a sodium vapor

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A parallel 1d3p Particle in Cell/Monte Carlo Collision (PIC/MCC) code was derived and applied for the investigation of the formation of photoplasma in sodium vapor. The effects of particle weighting and the Courant number on the computed plasma properties were examined, and the convergence of the numerical method with respect to these parameters was demonstrated. Simulations were carried out for the stepwise spatial profile of the resonant sodium atoms density. The basic plasma parameters (such as eedf, iedf, atomic and molecular ion and electron densities, and electric field and potential) were computed. The results of the PIC/MCC simulations were compared to those obtained from the fluid model. Simulations revealed a strong spatial non-uniformity in the electron density and the electric potential over the computational domain that provides evidence in favour of photovoltaic conversion of light energy into electrical energy.

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I. INTRODUCTION

Photoplasma is a partially ionized gas medium formed by the ionization of neutral gas by light radiation.1 Mohler and Boeckner, who observed an ionization that occurred in cesium vapor exposed to radiation, presented the first evidence of this phenomenon.2 The most commonly used method for the generation of photoplasma is based on the resonance absorption of the incident light. In this situation, the charged particles are mainly created in Penning and associative ionizations. Photoplasma is also often formed by laser-induced ionization of atoms by simultaneous or stepwise absorption of one or more photons of different frequencies, in general.1

Systematic studies of photoresonance plasma were started in 1967 by Morgulis, Korchevoi and Przhonskii,3 who generated the photoplasma of rather high density in cesium vapor irradiated by the cesium light sources. The ensuing investigations4,5 dealt mainly with the mechanisms and rates of processes induced by the excited atoms.

The photoplasma attracts attention due to its applications in the charge-transfer collision cross-section measurements, the production of isotopically pure elements, the purification of alloy, etc.1 Potentially promising application deals with the possibility of using photoplasma for the direct conversion of solar energy to electric energy.6 The first experimental evidence of this conversion was demonstrated in Ref. 7, where the photoplasma was created by heating cesium vapor with concentrated solar radiation.

The approaches that are based on the photovoltaic effect in photoplasma and intended to convert solar energy to electrical energy include the MHD generators,7 the thermionic converters,8 and the methods exploiting non-homogeneity of the plasma.9 In typical photoplasma devices,1 alkali metal (mainly Na and Cs) vapors are employed, because their resonance levels fall in the visible light spectrum that facilitates the use of light as an excitation source. For instance, in sodium vapor, the main absorption line is at 589 nm that corresponds to the 3s → 3p transition such that sodium atoms are excited from 3s2 1S1/2 to 3p2 3P3/2. An additional point that should be emphasized is that the alkali metals have a low-melting point.

A setup with a double-chamber plasma cell, considered recently in Refs. 10 and 11, tends to be relevant to the problem of conversion of solar energy to electric energy. The reason is that large spatial gradients of electron density and temperature, leading to an appreciable electric potential between the chambers, are formed in that system. The models in Refs. 10 and 11 are based on the fluid approximation, which is often not applicable and therefore, a kinetic analysis of the plasma parameters is required. However, the theoretical investigations of photovoltaic plasma converters presented so far are limited to the analytical models7 and simplified numerical models in one space dimension.9 In order to adequately understand the physics underlying this subject, we carried out a kinetic analysis of photoplasma. Specifically, we performed 1d3p parallel Particle in Cell/Monte Carlo Collision (PIC/MCC) simulations of photoplasma in sodium alkali metal vapor, exposed to light radiation.

The numerical method is summarized in Sec. II. The model and the parameter regime are described in Sec. III. Verification of the numerical model and code, and the analysis and discussions of the results are given in Sec. IV. Finally, conclusions are presented in Sec. V.

II. PIC/MCC METHOD

Particle in Cell/Monte Carlo Collision (PIC/MCC) is a numerical method that provides a way to perform a precise quantitative analysis of plasma.12 PIC/MCC employs a concept of superparticles, which represent a large number of real particles. This makes it possible to deal with a reasonably large number of particles in simulations. In addition, the use of a computational grid (“cells”) provides great ease in
calculating the electric and magnetic fields and interpolating these values to the particle positions.

In the PIC/MCC simulation (we consider the case of a one-dimensional coordinate space and the absence of the magnetic field), the particles are initially distributed randomly over the computational domain. Then, the charge density $\rho$ is evaluated at the grid nodes. Next, the electric field $E$ is calculated from the Poisson equation,

$$-\varepsilon_0 \frac{\partial^2 \varphi}{\partial x^2} = \rho. \quad (1)$$

Then, new positions and velocities of the particles are determined from the equations of motion, which, in the case of the leapfrog method, are obtained in the form

$$v_{k+1/2} = v_{k-1/2} + q \frac{E(x_k) \Delta t}{m}, \quad (2)$$

$$x_{k+1} = x_k + v_{k+1/2} \Delta t. \quad (3)$$

Here, $q$ is the particle charge, $m$ is the particle mass, $\Delta t$ is the time step, and the index $k$ is the time level. Finally, the effect of collisions of particles and the boundary conditions are taken into account by the Monte Carlo Collision method. To detect the type of collision, the collision probability,

$$P = 1 - \exp \left( -n \sigma_{\text{tot}} \Delta t \right) \quad (4)$$

is calculated, where $n$ is the neutral density, $\sigma_{\text{tot}}$ is the total cross-section, and $v$ is the particle velocity. The specific collision type is determined from the comparison of the ratio of the corresponding reaction probability to the probability (4) with the random number distributed uniformly between 0 and 1. Then, the algorithm advances to the next time step, and the cycle is repeated.

When solving the Poisson equation (1), one of the boundary points was grounded, and at the other point, the potential was determined from the surface charge accumulated at that boundary by using the condition

$$\varepsilon_0 \nabla \varphi \cdot \hat{n} = -\sigma, \quad (5)$$

where $\sigma$ is the surface charge density and $\hat{n}$ is the normal unit vector outward of the wall surface.

Within the PIC/MCC method, the grid size must be comparable with the Debye length, while the time step must be selected according to the Courant condition, and it must be of the range of the plasma oscillation period. In addition, to get statistically reasonable results, the number of superparticles per Debye length must be sufficiently high. For details of the method, see, e.g., Refs. 13 and 14.

In the present work, we modified our parallel 1d3v PIC/MCC numerical code, which has been developed in Ref. 15, and applied it to the analysis of photoplasma formation in sodium vapor exposed to light.

III. PLASMA-CHEMICAL MODEL AND PARAMETER REGIME

In the model, we took into account the elastic,16 excitation, and stepwise (from the resonance level) ionization collisions17 between the sodium atoms and the electrons (Fig. 1).

The reactions between the sodium atoms and ions include the isotropic scattering and backward scattering processes,18 with cross-sections of $4 \times 10^{-14}$ cm$^2$ and $2 \times 10^{-14}$ cm$^2$.

The superelastic collisions

$$\text{Na}(3p) + e \rightarrow \text{Na}(3s) + e \quad (6)$$

have a cross-section $^{19}$ of $20 \times 10^{-16}$ cm$^2$, through which the energy of the excited atoms is transferred to electrons.

Finally, the reactions which play a dominant role in the plasma formation $^{20,21}$ include the associative ionization with a cross-section of $4 \times 10^{-17}$ cm$^2$

$$\text{Na}(3p) + \text{Na}(3p) \rightarrow \text{Na}_2^+ + e, \quad (7)$$

excitation in inter-atomic collisions with a cross-section of $1 \times 10^{-15}$ cm$^2$

$$\text{Na}(3p) + \text{Na}(3p) \rightarrow \text{Na}(nl) + \text{Na}(3s), \quad (8)$$

and Penning ionization with a cross-section of $1 \times 10^{-12}$ cm$^2$

$$\text{Na}(nl) + \text{Na}(3p) \rightarrow \text{Na}^+ + \text{Na}(3s) + e. \quad (9)$$

According to Carre et al., $^{21}$ Na(5s) and Na(4p) atoms prevail in Penning ionization. For simplicity, we took into account Na(5s) only.

We considered such a situation where the light radiates a portion of the discharge cell such that the distribution of the excited sodium atoms over the cell is inhomogeneous. To imitate this, simulations were performed by taking a stepwise Na(3p) density distribution, with the density equal to $4.2 \times 10^{18}$ m$^{-3}$ for $0 < x < 0.2$ cm and absent outside this interval. Also, it is noteworthy that this situation, to a certain extent, imitates a dual-chamber setup, where an externally induced excitation is located within the smaller chamber of linear dimension $0 < x < 0.2$ cm, and the plasma generated expands to a larger chamber of size $0.2 < x < 1.0$ cm. As is shown in Refs. 10 and 11, large gradients of electron density and temperature, which lead to an appreciable electric potential

![FIG. 1. Electron cross-sections for elastic, excitation and stepwise ionization from the resonance level Na(3p) collisions in sodium used in the model.](image-url)
which provides a sufficiently high plasma parameter computed when the system settled down to a steady state, and time interval of 4 s. This corresponds to the “physical” energy distribution function (eedf) and the Na\(^+\) ion and electron mean energies (see Table II). With a decrease in the weighting (see Fig. 2) show that the computed results are qualitatively adequate already at weighting 4 \(\times 10^8\). As a result, the total number of superparticles in the system as well as the average number of superparticles per grid cell and the number \(N_D\) of superparticles per Debye length \(\lambda_D\) increased by about one order of magnitude. The parameters in the table were computed when the system settled down to a steady state, and \(N_D\) and \(\lambda_D\) were determined about the local maximum of the electron density. About 4 \(\times 10^6\) time steps were required to reach the steady state: this corresponds to the “physical” time interval of 4 \(\times 10^{-4}\) s.

Spatial distributions of the plasma parameters (the number density of Na\(^+\) ions, the electric potential, the electron energy distribution function (eedf) and the Na\(^+\) ion and electron mean energies) in Fig. 2 show that the location of the ionization sources at one part of the cell (0 < \(x < 0.2\) cm) and molecular Na\(^+\) ion density is much smaller compared to that of the atomic sodium ion density. The reason is that the rate of Penning ionization (9) far exceeds the rate of formation of molecular ions in associative ionization (7).

As can be seen from Fig. 4, which contains both the electron and the atomic Na\(^+\) and molecular Na\(^+\) ion density profiles, plasma in the volume is quasineutral and the molecular sodium ion density is much smaller compared to that of the atomic sodium ion density. The value of the Courant number \(C\) depends on the numerical method employed for the time integration. For example, for the explicit Eulerian leapfrog method\(^\text{23}\) employed in the present work, the Courant number is limited to 1.

In order to ensure the convergence of the numerical method with respect to the time step, we examined the effect of the Courant number on simulation results, by keeping the grid size \(\Delta x\) constant and reducing the time step \(\Delta t\). The velocity in the Courant condition was identified by the thermal velocity of electrons,\(^\text{23}\) \(v_{th} = \sqrt{2k_B T_e/m_e}\), where we employed the effective temperature \(T_e\) corresponding to highly energetic electron group [see Fig. (d)]. The effect of the Courant number on the total number of superelectrons, the number of superelectrons per Debye length, \(N_D\), and the ratios between \(w_{pe}\) and the time step \(\Delta t\), and between the Debye length \(\lambda_D\) and the grid size \(\Delta x\) are summarized in Table III. The effect of the Courant number on the spatial profiles of ions, the electric potential, and the eedf is shown in Fig. 3, which certainly implies the convergence of the numerical results with the time step refinement.
decay of the charged particles due to their ambipolar diffusion towards the walls lead to significant non-homogeneity in spatial distributions of the charged particle densities and the electric potential. This results in a noticeable potential drop (∼0.2 V) across the walls of the setup [see Fig. 2(b)]. This effect provides evidence in favour of photovoltaic conversion of light energy into electrical energy. Notice that the profile of the mean energy (the temperature) of electrons remains relatively uniform [Fig. 2(c)], because the magnitude of electron heat conductivity is greater than the system size, \[ \frac{\lambda T}{d} > L = 1 \text{ cm} \] (\( \lambda \) is the electron mean free path, and \( d = 2 m_e/M_e^2 \)). The ion temperature is rather uniform as well, and its magnitude, due to rapid energy exchange with the neutral atoms, is close to the neutral atom temperature. Correspondingly, the iedf is practically independent of the coordinate: the effect of the position on the 

<table>
<thead>
<tr>
<th>Courant number</th>
<th>Total number of super electrons ( N_D )</th>
<th>( w_{pe} )</th>
<th>( \Delta t )</th>
<th>( \Delta x/D_x )</th>
<th>( T_h (eV) )</th>
</tr>
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<tr>
<td>1</td>
<td>2600553</td>
<td>6475</td>
<td>0.88</td>
<td>0.67</td>
<td>0.28</td>
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<tr>
<td>2</td>
<td>2747504</td>
<td>6127</td>
<td>0.46</td>
<td>0.75</td>
<td>0.24</td>
</tr>
<tr>
<td>3</td>
<td>2721060</td>
<td>6068</td>
<td>0.23</td>
<td>0.75</td>
<td>0.24</td>
</tr>
</tbody>
</table>

FIG. 2. Effect of particle weighting on (a) the Na\(^+\) ion number density, (b) the electric potential, (c) Na\(^+\) ion and electron mean energies, and the eedf (d). The labels 1–4 for the weighting correspond to those in Table II. The panels (a) and (b) also contain profiles of the particle density and the potential obtained from the fluid model. \( T_h \) and \( T_l \) in panel (d) indicate the effective temperatures corresponding to high and low energetic electron groups. The parameters used in the simulations are summarized in Table I.
distribution function was found to be small. The uniformity in the ion mean energy is violated in the near-wall regions, where the potential drop accelerates the ions and leads to an abrupt jump in their mean energy profile [Fig. 2(c)].

As is illustrated in Fig. 5, the profiles of the eedf corresponding to the thermal range of the spectrum (formed by the bulk electrons) are relatively similar at different regions of the plasma. Their shape is Maxwellian, with the temperature approximately equal to 2/3 of their mean energy [see Fig. 2(c)]. An inflection point in the eedf is noticeable at 1 eV, where the slope of the distribution function changes greatly. This energy corresponds to the energy of electrons produced in Penning ionization [see Eq. (9)], which is the basic reaction in which the charged particles are created in the system. In the steady state, the rate of birth of the charged particles is equal to the rate of their ambipolar death at boundaries. Since fluxes of the ions and electrons to the walls must be equal to one another, the ambipolar flux of the ions towards the boundary is equal to the flux of electrons generated with an energy of 1 eV in reaction (9). Therefore, the wall potential (the potential drop between the location of the maximum particle concentration and the walls) must approximately correspond to this value of energy. This situation is analogous to the phenomenon of the occurrence of an abnormal potential jump, observed in the non-local current free plasma containing fast electrons, which equates the ion end electron fluxes towards the walls. Therefore, electrons with energies less than 1 eV are locked within the volume and undergo energy relaxation in elastic electron-atomic collisions that lead to the formation of the eedf in the region $e < 1$ eV. Electrons with energies $e > 1$ eV, in turn, diffuse freely towards the walls. This explains the occurrence of the inflection point in the eedf at about $e = 1$ eV.

D. Comparison to the fluid model

We performed a numerical analysis of the photoplasma parameters by employing a fluid model. This model is widely used in practice, because it is quite simple and provides a quick and easy way to estimate the plasma parameters. It is based on the diffusion-drift theory of gas discharge and includes the continuity equations for the electrons and positive sodium ions with number densities $n_e$ and $n_i$,

$$\frac{\partial n_e}{\partial t} + \frac{\partial}{\partial x} (n_e \mu_e E - D_e \frac{\partial n_e}{\partial x}) = S_e, \quad (10)$$

$$\frac{\partial n_i}{\partial t} + \frac{\partial}{\partial x} (n_i \mu_i E - D_i \frac{\partial n_i}{\partial x}) = S_i, \quad (11)$$

coupled to the Poisson equation for the electrostatic field,

$$\frac{\partial E}{\partial x} = \frac{e}{\varepsilon_0} (n_i - n_e), \quad E = -\frac{\partial \varphi}{\partial x}. \quad (12)$$

Here, $\varphi$ is the electric potential, $E$ is the electric field, $e$ is the electron unit charge, and $\varepsilon_0$ is the dielectric constant. The mobility and diffusion coefficients for electrons and ions $\mu_e$, $D_e$, and $\mu_i$, $D_i$ are related by the Einstein relation. The source terms in the continuity Eqs. (10) and (11) are defined by Penning ionization only, with the rate coefficient taken as $21.6 \times 10^{-9}$ cm$^3$/s. The parameters used in this model are summarized in Table IV.

<table>
<thead>
<tr>
<th>Species</th>
<th>$e$, Na$^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>System size (cm)</td>
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</tr>
<tr>
<td>Gas temperature (eV)</td>
<td>0.03</td>
</tr>
<tr>
<td>Electron temperature (eV)</td>
<td>0.3</td>
</tr>
<tr>
<td>$\mu_e$ (m$^2$/V s)</td>
<td>54</td>
</tr>
<tr>
<td>$\mu_i$ (m$^2$/V s)</td>
<td>18</td>
</tr>
</tbody>
</table>
from the fluid model, nevertheless, demonstrate qualitatively proper behaviour.

V. CONCLUSIONS

We carried out the Particle in Cell/Monte Carlo Collision (PIC/MCC) analysis of the problem of formation of photoplasma in the vapour of sodium. We modified and adapted our parallel 1d3v PIC/MCC code developed before in Ref. 15.

To ensure the numerical convergence and obtain adequate results, we first examined the effects of the particle weighting and of the Courant number on the plasma parameters. Numerical simulations were performed for the stepwise spatial profile for the resonant sodium atoms Na(3p). The basic discharge plasma parameters (such as the eedf, the iedf, the atomic and molecular ion and electron densities, the electric field and potential, and the mean energies of the particles) were computed. The results of the PIC/MCC simulations were compared to those obtained from the fluid model.

The computed results reveal strong non-uniformity in spatial distributions of the electron density and the electric potential across the computational domain that provides a notable electric potential between the walls of the plasma cell. This effect presents evidence for the feasibility of photovoltaic plasma converters.9

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