Au-II 282 nm segmented hollow-cathode laser-parametric studies and modeling

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Laser operation on the Au-II 282.3 nm ultraviolet transition is obtained using a high-voltage segmented hollow-cathode discharge tube. The metal vapor is produced by means of cathode sputtering. A small amount of argon is added to the helium buffer gas in order to achieve higher sputtering yield. Measurements of the laser power and small signal gain indicate that the optimal partial concentration of argon is in the range of 0.25%–0.75%. Quasi-continuous wave output power of 100 mW is obtained from a 34-cm-long active region while the highest small-signal gain is 52% m⁻¹. To explain the basic features of the laser operation we present a model of the segmented hollow-cathode discharge. All the discharge characteristics are calculated in a self-consistent way except the temperature of slow electrons. The trajectories of fast electrons emitted from the cathode are followed by Monte Carlo simulation. Rate equations of ion, metastable and metal atom densities are solved in the negative glow, while another Monte Carlo code is applied for the fast heavy particles in the cathode sheath. The spatial distribution of the gas temperature and the thermalization of sputtered metal atoms are calculated as well. The laser characteristics predicted by the model are in reasonable agreement with the experimental results. © 2002 American Institute of Physics. [DOI: 10.1063/1.1510172]

I. INTRODUCTION

Hollow-cathode discharges have been applied for decades as pumping sources of metal ion lasers. Different arrangements utilize thermal evaporation or cathode sputtering for metal vapor production. Unlike heated systems, the cathode sputtered laser tubes are usually easy to operate. Despite this fact (mostly due to their limited lifetime) no commercial sputtered lasers were available until very recently. However, as these low-cost metal ion lasers are capable of lasing in the deep UV region they are prospective light sources for UV Raman spectroscopy.

A. The helium-gold laser

Laser oscillation on the Au-II 282.3 nm (5d⁹7s-5d⁹6p) transition was reported first by Reid, McNeil, and Collins using a slotted hollow-cathode discharge. In the last 20 yrs various high-voltage hollow cathode constructions (such as hollow-anode cathode or helical cathode) were developed to improve the performance of cathode sputtered metal ion lasers. One of the newest types is the so-called segmented hollow-cathode (SHC) electrode arrangement, which has proven to be an outstanding pumping source for metal ion lasers. Earlier studies of the SHC Au-II 282.3 nm laser were published by Peard et al., more detailed parametric measurements are presented here. It is also to be mentioned that laser action on the Au-II 282 nm transition was recently obtained in radio frequency and also in heated pulsed positive column discharges.

The 7s levels of Au-II are populated by thermal energy charge transfer collisions between helium ions and ground state gold atoms. It is evident that for an efficient pumping of the laser a high density of both species must be present in the discharge. In cathode sputtered systems higher metal vapor density can be achieved by adding a small amount of a heavier noble gas, e.g., argon, to the helium buffer gas. In this case argon ions enhance the cathode sputtering. On the other hand, argon atoms—having low ionization potential and high ionization cross section—are easily ionized by electron impact. Therefore, with increasing argon concentration one can expect a lower ionization rate of helium. The final effect of the argon on the laser operation depends on the balance of the beneficial and disadvantageous processes mentioned above. As a result, an optimal argon concentration exists for which the best laser parameters are obtained.

B. Modeling of hollow-cathode laser discharges

The majority of glow-discharge modeling studies deal with the case of low current densities. Precise description of the discharge structure can be obtained using hybrid models that combine fluid description of slow ions and electrons with a particle (Monte Carlo type) simulation of fast plasma components (electrons, ions and atoms). This way the discharge structure and the electric parameters can be readily reproduced.

Compared to hybrid modeling, an alternative approach is usually chosen in simulations of high current laser discharges. Such a self-consistent model of a hollow-cathode...
helium–mercury laser was reported by Fetzer and Rocca.\textsuperscript{21} In their laser the metal vapor was produced by thermal evaporation. Comprehensive self-consistent modeling of cathode sputtered helium–copper laser discharges with different hollow-cathode configurations was carried out by Arslanbekov, Tobin, and Kudryavtsev.\textsuperscript{22,23} In these models the discharge volume is divided into the negative glow, which is assumed to be “field free,” and the cathode sheath, where the voltage drop occurs. In all cases a set of coupled rate equations is solved for the densities of ionic and neutral species in the negative glow. The major differences between the models are related to the description of fast electrons, which are responsible for the ionization of buffer gas atoms and the treatment of fast ions (and atoms) moving towards the cathode in the cathode fall region. In the model of Fetzer and Rocca\textsuperscript{21} the sheath is divided into a discrete set of equipotential cells. The cell width and the corresponding local electric field strength are derived from the Poisson’s equation. The energy distribution of the ion and electron fluxes are calculated within each cell (moving step by step towards and away from the cathode, respectively). Numerical solution of the Boltzmann equation is applied to obtain the energy distribution of energetic electrons in the negative glow, which is assumed to be homogeneous (zero dimensional). On the other hand, an integro-differential form of the Poisson’s equation is applied in the models of Arslanbekov and co-workers. Using the ion flux and the sheath thickness as a parameter, this equation gives a global description of the electric field in the cathode fall region. The distribution function of electrons is calculated analytically adapting the straight line trajectory and the continuous-slowing-down approximations. The analytical approach is also chosen to obtain fast ion and fast atom energy distribution functions in the cathode sheath, while the elastic collisions of ions are neglected.

In the present work the achievements of the above models are combined with Monte Carlo (particle) simulation of fast electrons and fast heavy particles. The method is based on the description of individual collisions of (discrete) particles, this way giving a realistic picture of elementary processes taking place in the plasma. The macroscopic characteristics are obtained by averaging over sufficiently large ensembles of species. Different elementary processes can easily be included in such models, provided that the appropriate cross sections are known. Using a Monte Carlo code for fast electrons we can obtain an improved image of the spatial distribution of ionization and excitation events compared to the straight line trajectory approximation. The resulting sources of ionic species (using our model) are not restricted to the segments bounded by the two cathodes, which gives a more realistic spatial distribution of charged particles. The Monte Carlo approach for the transport of fast particles through the sheath results in more precise energy distribution of particles bombarding the cathode as well. These energy distributions have a significant effect on plasma parameters. On the other hand, a prolonged computational time is needed for particle simulation programs—as compared to “macroscopic” or analytic approach represented by earlier models\textsuperscript{21,22}—to achieve sufficient statistics.

II. EXPERIMENT

Figure 1 shows the cross section of the SHC discharge used in our experiments. The discharge tube is formed by six identical discharge modules each 5.6 cm long with an inner diameter of 3.1 mm.\textsuperscript{24} The active areas of the cathodes were gold plated to a thickness of 30 \(\mu\)m. Although the excitation mechanism of the laser allows continuous wave operation, our experiments are carried out with pulsed excitation (0.5-ms-long rectangular current pulses at 1 Hz repetition rate) to avoid overheating and cathode erosion problems.

The 1.5-m-long optical cavity consists of two concave mirrors, each having 3 m radius of curvature. One of the mirrors has \(\sim 0\%\) nominal transmittance, while we use another \(\sim 0\%\) mirror for small signal gain measurement and a \(\sim 2\%\) transmittance mirror for the measurements of the laser power.

To determine the gain of the laser a calibrated loss element (a pair of contra-rotating quartz plates) is inserted into the resonator. Being aligned at the Brewster angle the plates do not effect the beam, except their absorption and scattering losses. The detailed description of the applied technique is given by Peard et al.\textsuperscript{11} The tube is divided into two parts—oscillator and amplifier—excited by two independent power supplies. First the current in the oscillator (comprising four discharge modules) is adjusted to bring the laser to threshold. This way the overall losses of the resonator are compensated. The desired current is then established in the amplifier (one discharge module) resulting in an increase of the output power. Finally, the loss element is adjusted to reestablish threshold operation. The gain per pass for the amplifier is then equal to the additional loss inserted by the quartz plates which can be calculated using the proper Fresnel formula.

As the modeling results depend sensitively on the temperature \(T_e\) of low-energy Maxwellian electrons in the negative glow (see Sec. III C), this quantity is determined experimentally in pure helium. It was shown by Hurt and Robertson\textsuperscript{25} that local thermal equilibrium (regulated by \(T_e\)) exists between the population of the \(n\Delta^2D\) (\(n\geq 8\)) levels of He in the negative glow of dc discharges. The existence of such an equilibrium depends also on the density of electrons, which is sufficiently high at our conditions.\textsuperscript{26} It follows that the electron temperature can be obtained by measuring intensity ratios of the lines originating from these states and terminated on the \(2p^3P\) level. The calculation procedure is similar to that given by Warner\textsuperscript{27} or Leigh.\textsuperscript{28} The spectrum of the discharge light is recorded using a PGS-2 monochromator equipped with a photomultiplier tube. Using 30 \(\mu\)m slits, a resolution of \(\Delta \lambda = 0.25\) Å is achieved. Measurements are carried out for constant linear current density \((I = 0.07\) A cm\(^{-1}\)) and different pressures. Unfortunately, the low intensity of the transitions belonging to \(n\geq 11\) and the near coincidence of some of the lines with Au-I and Au-II transitions makes the evaluation of \(T_e\) less accurate. Moreover, unidentified transitions belonging to residual gases (probably originating from the sealing epoxy) were found in the spectra.

III. THE MODEL

As mentioned earlier, in the model of the SHC discharge presented here the discharge volume is divided into the nega-
tive glow and the two cathode sheath regions that are shown in Fig. 1. The model consists of five interconnected blocks as illustrated in the flow chart of Fig. 2. In the first two blocks the motion of fast electrons is modeled by Monte Carlo simulation and the thermalization of sputtered metal atoms is calculated. The source functions of ions, metastables and sputtered metal atoms are obtained, which serve as an input for the block of the negative glow region. Solving a set of coupled rate equations the spatial distributions of different species are calculated here. Next, a heavy particle Monte Carlo code is applied to follow the trajectories of ions and fast neutrals in the cathode fall region and the Poisson’s equation is solved to obtain the electric field distribution in the sheath. Finally, the spatial distribution of the buffer gas temperature is calculated. All the blocks are run iteratively until convergence is reached.

The discharge is assumed to be longitudinally uniform. The dimensions of the different variables in the model are taken as follows. The electric field is homogeneous in the tangential direction, and only radial field is considered. The motion of fast electrons is followed in three dimensions, however, as we assume uniform conditions along the tube axis, the resulting source functions of ions and metastables are reduced to be two-dimensional ($r, \varphi$) values. The same applies for the thermalization of sputtered metal atoms. The rate equations describing the negative glow are solved in two dimensions ($r, \varphi$). Fast ions and atoms are traced in three dimensions in the heavy particle Monte Carlo code, and the resulting charge density is averaged for each radial position in the sheath so that a one-dimensional ($r$) Poisson equation can be solved. Finally, the flux of particles hitting the cathode is averaged across the cathode area, and the spatial distribution of emitted electrons and sputtered metal atoms is taken to be homogeneous for the whole cathode surface. In the following we give a detailed description of each part of the model.

A. Fast electrons

Bombardment of cathode by ions, metastables and fast neutrals results in emission of primary electrons that are accelerated towards the tube center by the electric field. These electrons—being responsible for the ionization of buffer gas atoms—play an important role in the self-sustainment of the discharge. Secondary electrons are created in ionizing collisions, this way giving rise to electron avalanches. For description of fast electrons the Monte Carlo codes published in Refs. 20 and 29 are modified to fit the conditions of the SHC discharge. It is assumed that primary electrons are emitted uniformly from the whole cathode surface with initial energy between 0 and 5 eV (chosen randomly) and with an initial velocity vector that is perpendicular to the cathode. The motion of primary and secondary electrons is traced until they are absorbed on the anode surface or—due to the energy losses in inelastic collisions—their total energy drops below the ionization potential of the buffer gas. Elastic scattering of electrons, electron impact ionization and excitation of helium, argon and gold is taken into account as well as the reflection of electrons from the anode. Between two collisions the equation of motion is integrated continually. The position of the next collision can be obtained from

$$\int_{s_0}^{s_1} \sum_k n_k \sigma_k(e(s))ds = -\ln(1-R_{01}).$$

(1)

Here $n_k$ is the density of neutral particles (index $k$ being the type), $\sigma_k(e)$ is the sum of the energy-dependent cross sections of the collisions between electrons and the $k$th particles, $R_{01}$ is a random number uniformly distributed in the [0,1] interval, $s$ is the curvilinear coordinate and $s_0$ and $s_1$ denote...
the position of the previous and next collision, respectively.\textsuperscript{30} The type of the collision that occurs after the “free flight” is chosen randomly. The probabilities of different processes are obtained from the cross sections belonging to the actual electron energy. During the elastic scattering process, which is assumed to be anisotropic (the form of the differential cross section is given by Birdsall\textsuperscript{31}) the energy loss of electrons is neglected. In the case of electron impact excitation the electron loses a random energy between the gas atom’s first excited level and its ionization potential. In the ionization process the probability distributions of energies and scattering angles of the electrons depend on the energy of the incident electron.\textsuperscript{30,32,33} The cross sections of electron collisions with helium atoms are taken from Ref. 29, data given in Ref. 34 are used for argon. The portion of electron collisions resulting in helium metastable atoms is evaluated from the sum of cross sections belonging to the electron-impact excitation of He\textsuperscript{1}S and He\textsuperscript{3}S levels taken from the Siglo database.\textsuperscript{35} For the collisions of electrons with gold atoms, we use ionization cross section published in Ref. 36, excitation cross sections given in Ref. 37 and the cross section of elastic scattering reported in Ref. 38.

As only radial electric field is considered in the model, a potential jump occurs at the cathode–anode transition region of the sheath. Electrons passing through this wall gain the energy that belongs to the given radial position.

After emission of a sufficient number of electrons (typically 5000 in each iteration step of the model) the spatial distributions of the ionic sources \( S_i(r, \phi) \) are obtained, while the actual electron current on the cathode (see Sec. III D) is used for normalization. Following the approach of Arslanbekov,\textsuperscript{22} an effective helium metastable state is introduced in the model, the source of which is also calculated here.

### B. Thermalization of sputtered gold atoms

Slowing down of sputtered particles is treated in accordance with the theory of velocity proportional stopping given by Gras-Marti and Valles-Abarca.\textsuperscript{39} The energy spectrum of emitted particles is approximated by

\[
\Psi(E, \theta) = \frac{2UE}{(E+U)^3} \cos \theta, \tag{2}
\]

where \( \Psi \) is the normalized extracted flux of particles having energy \( E \) in the direction \( \theta \). The maximum of the above distribution occurs at the energy \( U/2 \). Experimental results of Stuart, Wehner, and Anderson\textsuperscript{40,41} indicate that Eq. (2) is valid only to a certain extent. For instance, it was found that the most probable energy \( E_{mp} \) (corresponding to \( U/2 \)) depends on the type, but does not depend on the energy \( E \) of impinging species. On the contrary, the average energy \( \bar{E} \) of emitted particles increases with the energy of bombarding atoms. To account for this fact—still keeping the advantage of the analytical expression (2)—an energy dependent \( U(\varepsilon) \) is introduced in the model. Data of \( E_{mp} \) measured for krypton ions bombarding gold surface and the shape of \( \bar{E}(\varepsilon) \) given in Refs. 40 and 41 for sputtering of copper are used for estimation of \( U(\varepsilon)/2 \). For the sake of simplicity in the calculations we only use the values of \( \langle U_p(\varepsilon)/2 \rangle \) averaged over the flux energy distributions \( \Phi_p(\varepsilon) \) of different impinging species \( (p = \text{He}^+, \text{Ar}^+, \text{Au}^+, \text{He}, \text{Ar}, \text{Au}) \). \( \Phi_p(\varepsilon) \) is obtained from the cathode sheath part of the model, see Sec. III D. When compared to sputtering by krypton ions the energy of emitted metal atoms is taken to be 20% smaller in the case of argon ions,\textsuperscript{41} while 60% smaller and 60% higher values are used for helium and gold ions, respectively.\textsuperscript{40} It is assumed that there is no difference between bombarding by ions and fast atoms of the same type and energy.

The final goal is to determine the source function of thermalized metal atoms \( S_{\text{Au}} \) on the grid that is used in the model of the negative glow region. The calculation procedure is as follows. First, the flux of sputtered particles originating from elemental cells homogeneously distributed over the cathode surface is analyzed. The motion of metal atoms emitted from different positions (with different energies \( E \), emission and azimuth angles \( \theta \) and \( \phi \)) is traced assuming the straight line trajectory and velocity proportional slowing down approximations. The energy loss of the particles is calculated from

\[
\frac{dE}{dt} = -2/A \sqrt{E}, \tag{3}
\]

where the parameter \( A \) is inversely proportional to the density of the stopping gas and is given in Ref. 39. As the position of thermalized atoms is obtained (at \( E=0 \)), the value of the metal source function is increased proportionally to Eq. (2) in the nearest grid point. Finally, the contributions of different sputtering species are normalized according to their fluxes onto the cathode. The total flux of metal atoms is given by

\[
\Phi_{s}^\text{tot} = \sum_p \int_{e} \zeta_p(\varepsilon) \Phi_p(\varepsilon) d\varepsilon. \tag{4}
\]

The sputtering coefficients \( \zeta_i \) are collected by Matsunami and co-workers.\textsuperscript{32} Again, the sputtering yields of fast particles and corresponding ions are assumed to be the same.

### C. Negative glow

The densities of helium, argon and gold ions, helium metastables and neutral gold atoms are calculated in the field free negative glow region solving a set of two-dimensional coupled rate equations

\[
\frac{\partial \text{[He}^+\text{]}}{\partial t} = \nabla \cdot (D \text{[He}^+\text{]} \nabla \text{[He}^+\text{]} + S_{\text{He}^+} - \alpha_{\text{He}^+} \times \text{[He}^+\text{]} n_i^2 - k_2^\text{He}^+[\text{He}^+][\text{Ar}] - 2k_3^\text{He}^+[\text{He}^+][\text{Ar}] + 2k_4^\text{He}^+[\text{He}^+]^2,
\]

\[
\frac{\partial \text{[Ar}^+\text{]}}{\partial t} = \nabla \cdot (D \text{[Ar}^+\text{]} \nabla \text{[Ar}^+\text{]} + S_{\text{Ar}^+} - \alpha_{\text{Ar}^+} n_i^2 + k_5^\text{He}^+[\text{He}^+][\text{Ar}] + k_6^\text{He}^+[\text{He}^+][\text{Ar}] - k_7^\text{Ar}^+[\text{Ar}^+] [\text{Au}].
\]
Only the significant elementary processes are taken into account. The results of the “fast electron—Monte Carlo” and “metal thermalization” blocks are utilized as the source terms of helium and argon ions, helium metastables and gold atoms. The electron impact ionization of gold is neglected as compared to the charge transfer and Penning ionization of gold neutrals.\(^1\) The rate coefficient \(k_1\) of the charge transfer reaction between \(\text{He}^+\) and \(\text{Au}\) is calculated from the cross section that was estimated in Ref. 44. \(k_1\) is given in the compilation of Ikezoe et al.\(^1\) To our knowledge, cross sections for charge transfer between argon ions and gold atoms are not available in the literature (see discussion in Ref. 46) thus, \(k_1\) is approximated by the value that belongs to copper atoms.\(^2\) Due to the lack of data on the Penning ionization of gold by helium metastables the cross section of this process is approximated by the value published for the helium–mercury system.\(^3\) Rate constants of Penning ionization between helium metastables and argon atoms \(k_2\) and between two metastables \(k_3\) are taken from Ref. 22 as well as the rate of metastable quenching by thermal electron impact \(k_\text{eq}\). Based on the shape of (low energy) electron energy distribution function given by Arslanbekov and Kudryavtsev\(^4\) and the results of our fast electron Monte Carlo code we conclude that electron impact ionization from metastable helium state (for cross section see Ref. 48) can be neglected at typical discharge conditions in the model (see later).

The rate coefficients of collisional-radiative recombination \(\alpha_r\) (taken for helium and estimated for argon and gold on the basis of Ref. 49) depend strongly on the temperature of bulk electrons. For example \(\alpha_{\text{He}}=4.0 \times 10^{-19} \ T_e^{-5} \text{ cm}^6 \text{s}^{-1}\). Because of the high densities of charged particles, recombination plays an important role in hollow-cathode laser discharges. The energy balance of the low-energy, Maxwellian electrons was studied in detail in Refs. 50 and 51. Instead of the complex calculation procedure in our model the temperature of electrons \(T_e\) is used as a fitting parameter. Its value is adjusted to reproduce the measured voltage current characteristics. This approach is confirmed by our experimental results in the case of pure helium discharge. In the next section we give a more detailed discussion of the fitting procedure in connection with the electron yields of ions impinging on the cathode, which is the other parameter that may be fitted in glow discharge models.\(^5\)

The diffusion coefficient of ionic species \(D^+\) is the ambipolar diffusion coefficient and is calculated from the Einstein relation. Mobility data published in Ref. 52 are used for helium and argon, while the mobility of gold ions at room temperature is interpolated from Ref. 53. The temperature dependence is evaluated using the method given by McDaniel.\(^5\) Preliminary absorption measurements carried out in the discharge afterglow (using the method of Rusinov, Paeva, and Blagoev\(^5\)) provided us with the thermal diffusion coefficient of gold atoms in helium \(D_\text{Au}=340 \text{ cm}^2\text{s}^{-1}\text{Torr}\) and the coefficient of their reflection from the walls \(\rho=0.8\).

The following boundary conditions are used while solving Eq. (5). The density of ionic species and metastable atoms is set to zero at the walls. The negative glow—cathode sheath boundary is assumed to be a “perfect absorber” for the ions. This approximation is justified by the fact that the typical ion densities in the sheath are two orders of magnitude smaller as compared to the tube center due to the accelerating electric field. To account for the reflection of gold atoms from the walls we use the boundary condition given in Ref. 54.

The partial differential Eqs. (5) are solved using a finite difference technique on a two-dimensional \((r, \varphi)\) grid applying the Crank–Nicolson scheme. Among others, the densities of helium ions and gold atoms are obtained, the product of which determines the pumping rate of the upper laser level. As no potential barrier is assumed for the ions in front of the anodes (see also discussion of Arslanbekov, Tobin, and Kudryavtsev\(^2\) and results of Kutasi and Donko\(^2\)) ions diffusing towards the anodes are lost from the discharge model. The flux of ions entering the cathode sheath is defined by the derivatives of the ionic densities at the negative glow—cathode sheath boundary (NG-CS) boundary.

**D. Cathode sheath—heavy particle Monte Carlo model**

Ions passing through the NG-CS boundary or created in the sheath due to ionizing collisions are driven towards the cathode by the electric field. The distribution of the field is in turn determined by the presence of charged particles. The electrons, being much faster than the ions, do not contribute significantly to the space charge, thus their effect can be neglected. The electric field in the sheath is assumed to be homogeneous in tangential direction, and only radial field is considered in the model. The motion of ions and atoms is followed in three dimensions, primary ions originating from the negative glow start with a homogeneous distribution from the central part of the NG-CS boundary (here the radius \(r=r_0=\text{const}\)). Typically, the flux of ions entering the sheath through the anode sides of the NG-CS boundary represents \(\sim 10\%\) of the overall ion flux coming from the negative glow and is neglected in the model.

Trajectories of fast heavy particles in the sheath are followed by Monte Carlo simulation based on the same principles as used in the block of fast electrons. Ions are only traced until they reach the cathode. During the collision on...
the wall they lose their charge and are reflected as neutrals. Opposite to the electrons (Sec. III A), ions reaching the cathode–anode transition region of the sheath are reflected from the potential barrier. On the other hand, fast neutrals (playing an important role in cathode processes) are also created during charge transfer and elastic collisions of ions with buffer gas atoms. They are followed until their energy drops below 9(3/2)kTg, which is the threshold used to distinguish between the groups of “thermalized” and fast particles (see also Sec. III E). Neutrals hitting the electrodes are reflected from the surface and remain in the model. The fraction of energy that is lost in the collisions of particles on the electrode surface depends on their initial energy and is calculated according to the formula given by Winters and co-workers. The direction of reflected particles is assumed to have a cosine distribution.

Collisional processes included in the sheath model are summarized in Table I. The references indicate sources of cross sections (or sources of rate coefficients used for the estimation of cross sections). The collisions labeled as elastic scattering are taken to be isotropic in the center-of-mass system using the following cross sections. Viscosity cross section, multiplied by 1.5 are used for the scattering of fast helium atoms on helium, diffusion cross sections are taken for collisions of argon ions and atoms with helium. As the relative densities of argon and gold atoms are low, the collisions of fast neutrals with these species are fully neglected. The isotropic part of the differential cross sections (in center of mass) is neglected for the collisions of argon and gold ions with their parent atoms, and only the symmetric charge transfer processes are taken into account. Thermal energy rate coefficient (see Sec. III C) is used for estimation of charge transfer cross section of helium ions with gold atoms. Due to the lack of data the cross sections of charge transfer collisions of argon and gold ions with gold atoms are replaced with data belonging to charge transfer reaction of argon ions and copper atoms and the symmetric charge transfer of mercury ions and atoms, respectively. Ionization collisions are considered to be isotropic in the center-of-mass system. In our model gold ions can lose energy only by the symmetric charge transfer process; the effect of their scattering on the helium background is not taken into account.

Knowing the trajectories of different ions in the sheath, the charge density is calculated as a function of the radial position. For that the sheath is divided into 300 equidistant parts (separated by dr = const). For each ion we calculate the integrated time $t_{ij}^\text{on}$ that it spends in the $j$th part of the sheath. The total charge in the $j$th part is given by $Q_j = e \sum_{i,j} n_j^i t_{ij}^\text{on}$, where $dt$ is the basic iteration time used in the model (usually $dt$ is few times $10^{-15}$ s). The new distribution of the radial electric field is obtained from the Poisson equation:

$$E_j = \frac{\sum_{i,j} 2Q_i}{\varepsilon_0 \pi r_j^2}.$$  

This formulation is consistent with the assumptions of the model (there is only radial field in the sheath, there is no electric field in the negative glow). The potential distribution is obtained next:

$$U_j = \sum_{j=1}^{n-1} dr (E_j + E_{j-1})/2,$$

where $n$ is the number of parts the sheath is divided into. The length of the sheath $l$ is then modified:

$$\ell' = \ell (U/V)^{1/2},$$

where $U$ is the overall potential drop and $V$ is the voltage applied on the tube. The value of the factor ($n = 1/7$) is chosen empirically to ensure fast convergence towards the stationary state (while keeping other discharge parameters constant).

At the cathode surface the energy distributions of fast heavy particle fluxes $\Phi_p(\epsilon)$ are calculated. Electrons are assumed to be emitted from the cathode due to the impact of ions, fast neutrals and metastables

$$\Phi_{e}^{\text{tot}} = \sum_p \int_\epsilon \gamma_p(\epsilon) \Phi_p(\epsilon) d\epsilon + \gamma_m \Phi_{m}^{\text{tot}}.$$  

(6)

It is known that the values of electron emission coefficients $\gamma$ play an important role in glow discharge models. Unfortunately, $\gamma$ may change significantly depending on the actual condition of the surface. In many previous models the energy dependence of these coefficients was neglected. However, experimental values of $\gamma$ for helium ions and neutrals impinging on contaminated gold surface indicate a relatively strong energy dependence with zero energy limit of 0.23 (electron/incoming ion) for the ions. On the other hand, the energy dependence of the emission coefficient obtained at clean surface is less pronounced and the zero energy limit reported for ions is only 0.15 in this case. The difference in $\gamma$ between the dirty and clean surface at 500 eV is more than a factor of 3. We believe that in our sputtering discharge the $\gamma$ is somewhere between these two extreme cases. To eliminate this uncertainty the electron yields can be used as a fitting parameter in order to reproduce the measured voltage–current characteristics of the discharge, as was done by Kutasi and Donkó. In the recent model we apply the alternative way of fitting the temperature of slow electrons. For each discharge condition $T_e$ is adjusted to reproduce the measured voltage–current characteristics. As the temperature of electrons was also obtained experimentally for the case of pure helium discharge, we can use this additional information to reduce the uncertainty of the electron yields. We found that the fitted electron temperature values overlap with our experimental data (see Fig. 4) when the electron emission coefficients of helium ions are taken to be 0.67 times the ones reported for contaminated electrodes. The limitations of our assumptions are discussed later. The emission yield of metastables is taken to be the same as for helium ions in the

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<tr>
<th>Process</th>
<th>Refs.</th>
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<tr>
<td>$\text{He}^+ + \text{He} \rightarrow \text{He}^0 + \text{He}^0$</td>
<td>Isotropic scattering</td>
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<td>$\text{He}^+ + \text{He} \rightarrow \text{He}^0 + \text{He}^0$</td>
<td>Charge transfer</td>
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<td>$\text{He}^0 + \text{He} \rightarrow \text{He}^0 + \text{He}^0 + e$</td>
<td>Ionization</td>
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<td>$\text{He}^+ + \text{He} \rightarrow \text{He}^0 + \text{He}^0$</td>
<td>Elastic scattering</td>
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<td>$\text{He}^+ + \text{Ar} \rightarrow \text{He}^+ + \text{Ar}^+$</td>
<td>Charge transfer</td>
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<tr>
<td>$\text{He}^+ + \text{Au} \rightarrow \text{He}^+ + \text{Au}^+$</td>
<td>Charge transfer</td>
</tr>
<tr>
<td>$\text{Ar}^+ + \text{He} \rightarrow \text{Ar}^+ + \text{He}^0$</td>
<td>Elastic scattering</td>
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<td>$\text{Ar}^+ + \text{Au} \rightarrow \text{Ar}^+ + \text{Au}^+$</td>
<td>Charge transfer</td>
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<tr>
<td>$\text{Au}^+ + \text{Au} \rightarrow \text{Au}^0 + \text{Au}^0$</td>
<td>Charge transfer</td>
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zero-energy limit.\textsuperscript{63} Experimental data for argon ions and neutrals can be found in the compilation of Phelps and Petrovic.\textsuperscript{63} A constant value of $\gamma=0.06$ is taken for argon ions and the effect of argon neutrals is neglected in the model. We assume that no electrons are emitted by gold ions and neutrals.\textsuperscript{64}

Finally, the new value of the discharge current is obtained from the sum of the ionic and electron fluxes.

**E. Gas temperature**

The distribution of gas temperature $T_g$ in the tube is determined from the equation of heat conduction

$$\nabla \cdot [\lambda_g(T_g) \nabla T_g] = H_{\text{HP}} + H_{\text{SP}}. \quad (7)$$

Here $\lambda(T_g)$ is the temperature dependent heat conductivity of the buffer gas, while the right side of Eq. (7) represents the source terms. Only heating by collisions included in the heavy particle Monte Carlo model ($H_{\text{HP}}$) and heating due to the thermalization of sputtered atoms ($H_{\text{SP}}$) are taken into account. The effect of electrons is neglected.\textsuperscript{22} We follow the method published by Bogarts, Gibbels, and Serikov\textsuperscript{65} to evaluate $H_{\text{HP}}$. A threshold energy $E_{\text{thr}}=9(3/2)kT_g$ is applied to separate the groups of thermal and fast heavy particles. The source term is calculated as follows. First, $3/2kT_g$ is subtracted from the thermal source in each momentum transfer collision of fast particles with buffer gas atoms. Second, whenever a slow neutral with an energy less than the threshold is produced during a collision process, its energy is added to the source. The heating effect of sputtered metal atoms at different positions is taken as it follows from Eq. (3).

The wall temperature is assumed to be 350 K, the temperature jump occurring at the walls is treated in the same way as given in Ref. 23. The complete temperature distribution is utilized in the fast electron, sputtered metal thermalization and “negative glow” parts of the model, temperature averaged over the sheath at a given radius is used in the “cathode sheath” block.

**IV. RESULTS AND DISCUSSION**

In this section both experimental and modeling results are discussed and compared in a parallel way.

**A. Discharge characteristics**

1. **Discharge voltage and electron temperature**

The dependence of the discharge voltage on the argon percentage was measured for different buffer gas pressures. Data obtained at constant linear current density $j=0.07$ A cm$^{-1}$ are shown in Fig. 3 as a function of pressure. The curve belonging to pure helium shows a minimum, which is typical to hollow-cathode discharges.\textsuperscript{7} In general, the voltage is higher as compared to classical (cylindrical or slotted) hollow cathode arrangements, which is due to the increased losses of charged particles (ions and fast electrons) on the anodes. A further increase of the discharge voltage occurs when a small amount of argon (1%) is added to the buffer gas. At higher buffer gas pressures the voltage drops as the argon percentage is increased above 1%. This behavior differs from that observed in the SHC copper discharge, where a rising percentage of argon results in a monotonous decrease of the voltage.\textsuperscript{28}

The results of our model indicate that for the case of He+1% Ar mixture (at 20 mbar, 0.07 A cm$^{-1}$) 13% and 18% of the ionic current is carried to the cathode by argon and gold ions, respectively. Due to the higher secondary electron emission coefficient of helium ions the contribution of argon ions to the production of secondary electrons is only 4%; no electrons are emitted by gold ions in the model. It follows that the ratio of the electron to ion current on the cathode drops when argon is added into helium making the discharge less effective. However, we found that this effect alone cannot explain the increase of the voltage when argon is added to helium.

As mentioned earlier, the temperature of slow electrons is used as a fitting parameter in the model. Its value is set in a way that the measured voltage—current characteristics of the discharge are reproduced. Resulting data are shown in Fig. 4. It can be seen that—with the experimental errors—data measured spectroscopically in pure helium are in reasonable agreement with the predictions of the calculations.

This was achieved by the adjustment of the electron yields of helium ions and atoms as described in Sec. III D. For the estimation of the net effect of the argon admixture on the electron temperature self-consistent models are needed.\textsuperscript{50} Our results indicate a decrease of $T_e$ as more argon is added to the buffer gas. Coulomb collisions between slow and superthermal (<20 eV) electrons are believed to be the primary energy gain mechanism for cold electrons.\textsuperscript{51} The density of superthermal electrons may decrease at higher argon concentrations due to the inelastic collisions with argon and gold atoms. Thus, one can expect lower electron temperatures as argon is added to the buffer gas. On the other hand, more ions are lost by collisional-radiative recombination at lower electron temperatures, which can explain the increase...
of the discharge voltage. The model shows a relatively strong dependence of the voltage on the electron temperature. Increasing $T_e$ arbitrarily by 10% while keeping the voltage constant results in an increase of 17% in the discharge current (applies for a 20 mbar, 0.07 A cm$^{-2}$ discharge with 1% of argon in helium).

Because of the arcing tendency of the discharge in our latter measurements we had to restrict our investigation to the range of buffer gas pressure between 12.5 and 20 mbar (see also Ref. 7).

2. Negative glow

The negative glow part of the model provides us with the spatial distribution of different species that are included in Eq. (5). Typical data for helium ions and gold atoms are shown in Fig. 5. The highest He$^+$ values are located at the tube axis. For the conditions of Fig. 5 the losses of helium ions are distributed between the different channels as follows. Collisional radiative recombination represents 51%, charge transfer to argon and gold atoms represents 20%, while 29% of ions is lost from the volume by diffusion to the walls.

The highest density of gold atoms is in the front of the cathode surface [Fig. 5(b)], showing that the major part of sputtered atoms is thermalized on the first few tenths of a millimeter. They are reaching the tube center by diffusive motion. The two off-axis maxima of the distribution are connected to the cathodes, which may be the main reason for better performance of SHC metal ion lasers when the tube diameter is reduced (see results of Szalai et al.$^{66}$). The dependence of the average metal density on the concentrations of argon is shown in Fig. 6. Our data are in good qualitative agreement with those reported for SHC copper discharges.$^{9,67}$

At conditions of Fig. 5 the electron density at the tube center is $1 \times 10^{14}$ cm$^{-3}$, the corresponding density of metastable helium atoms is $4 \times 10^{12}$ cm$^{-3}$. Sixty-eight percent of primary ions followed by the Monte Carlo model in the cathode sheath originates from the NG-CS boundary. The remaining part is created during ionizing collisions of electrons in the sheath. This is different from what is obtained for low current plane-parallel discharges$^{68}$ where the ion flux at the NG-CS boundary represents only 30% of the total ion flux at the cathode. Consequently, in our case the discharge charac-
teristics are more sensitive to the density of ions in the negative glow, which favors the usage of the electron temperature as a fitting parameter.

3. Cathode sheath

The radial dependence of the electric field computed for the cathode fall region is shown in Fig. 7(a). The curving down part of the electric field near the negative glow boundary may be affected by the rough assumption of sharp borderline between the cathode sheath and the negative glow. However, similar tendency was found in the measurements reported by Lee et al. 69 and also in the modeling results of Fetzer and Rocca. 21 Changes due to the presence of argon are not pronounced, and the sheath becomes somewhat wider as voltage increases at higher percentage of argon. Higher space charge density and narrowing of the sheath corresponds to higher pressures and discharge currents.

Flux energy distributions of heavy particles bombarding the cathode are included in Fig. 7(b). There is a pronounced difference between distributions belonging to different ionic species. As helium ions undergo numerous symmetric charge transfer reactions in the sheath their energies are relatively small at the cathode. Consequently, the flux of fast helium neutrals is large. The maximum of the argon ion flux energy distribution is shifted towards higher energies, which makes the sputtering efficiency of argon ions high. Most of the gold ions reach the cathode without energy loss in the cathode sheath.

4. Gas temperature

According to computing results the temperature of the buffer gas averaged over the discharge volume changes in the range of 420–550 K for the conditions investigated in this article. For the set of parameters given in Fig. 5 the contribution of sputtered metal atoms to the total deposited power represents only 8%. The major part of gas heating belongs to heavy particle avalanches gaining energy in the sheath. From that 76% is due to fast neutrals reflected from the cathode. The regions of the highest temperature are found in front of the cathodes.

B. Laser characteristics

1. Small-signal gain

The small-signal gain is proportional both to the gold atom and helium ion densities. We neglect additional effects (such as the variation of the line broadening or the excitation of the lower laser level) and calculate a relative value of the small signal gain at each position as the product of He$^+$ and Au densities. The radial dependence of the gain along the lines connecting the middle of the opposite cathodes (and anodes) is shown in Fig. 8(a). The qualitative agreement with

FIG. 6. The dependence of the calculated average gold atom density on the argon concentration. The linear current density is 0.07 A cm$^{-1}$.

FIG. 7. (a) The electric field distribution in the cathode sheath at different conditions. (b) Flux energy distribution of heavy particles impinging on the cathode obtained at conditions given in the caption of Fig. 5.
experimental data measured for a 780.8 nm SHC copper ion laser having an inner diameter of 8 mm is very good.

The “average” small-signal gain—corresponding to recent measurements—is calculated from the gain profile weighted with a Gaussian function that belongs to the TEM$_{00}$ mode of the resonator. As the width of this mode is a few times smaller than the diameter of the tube, and the resonator was optimized in each measurement for the highest gain, the axis of the TEM$_{00}$ mode is placed to the position of the maximal gain in the calculations. Both the measured and calculated dependence of the small-signal gain on argon concentration is shown in Fig. 9. For higher pressures a broad maximum of the gain exists around 0.75% of argon. The optimal argon concentration is shifted towards higher values as pressure decreases. Our modeling results can reproduce the major features of this behavior. The differences may be attributed to the uncertainties of the data set used as an input of the model and also to the errors introduced by rough approximations applied both for the discharge structure and the gain. Also at higher pressures the effect of molecular ions (not included in the model) might start to play an important role. It is noted that modeling results represent steady-state conditions, whereas during the 0.5-ms-long discharge pulses applied in the experiment the pressure could not fully reach equilibrium in the long and narrow tube (see detailed discussion in our earlier article). This effect can also lead to some differences between the calculated and measured data.

Current dependence [Fig. 8(b)] was measured at optimal conditions up to 1 A applied to the amplifier module. This way small-signal gain of 52% m$^{-1}$ was reached. Steeper rise of the gain is predicted by the calculation as the current...
increases. However, the general shape of the measured curve is reproduced. The calculated temperature of electrons decreases with higher currents. The same tendency was found experimentally in SHC copper discharge operating at 14 mbar of helium with 4% argon mixture, however, the modeling results of Arslanbekov and co-workers show an opposite behavior. Our model fails to work at currents exceeding 0.11 A cm\(^{-2}\) where the discharge voltage is higher than 780 V. At these conditions the temperature of electrons required to regulate the current drops rapidly. We believe that this is due to the values of electron yields of helium ions and atoms at the cathode that are most probably overestimated at high energies. Consequently, the production of ions by electron impact ionization is too high and the collisional radiative recombination (strongly depending on \(T_e\)) cannot compensate for the increased rate of ionization.

2. Output power

Figure 10 shows the output power at different pressures as a function of the argon percentage. Data belong to the linear current density of 0.12 A cm\(^{-1}\). The transmittance of the output coupler (2%) was not optimized. It can be seen that, compared to the small-signal gain measurements, the optimal argon concentration at 20 mbar is shifted towards lower values.

V. CONCLUSIONS

The dependence of the 282 nm Au-II SHC laser operation on the argon added into the helium buffer gas was studied at different pressures. Optimizing the discharge parameters, maximal small-signal gain of 52% m\(^{-1}\) and quasicontinuous output power of 100 mW was reached.

In the model of the SHC sputtering discharge presented here Monte Carlo description of fast particle motion (electrons, ions and atoms) is combined with a fluid model of slow ions, metastables and metal atoms. We have shown that in the high current hollow-cathode laser discharge models the electron temperature can be used as a fitting parameter, so that the measured voltage–current characteristics are reproduced. However, the results may strongly depend on the electron emission coefficients of the particles bombarding the cathode, which represent the most uncertain input parameters in our case. The calculated data are in reasonable agreement with the measurements.

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