DOE/ER/13727--T3

FINAL REPORT

DOE Grant DE-FG03-87ER13727

Dynamics of Electronegative Plasmas for Materials Processing

Allan J. Lichtenberg and *Michael A. Lieberman*
Department of Electrical Engineering and Computer Sciences
and the Electronics Research Laboratory
University of California, Berkeley, CA 94720-1774

I Abstract

The purpose of this project is to study the equilibrium particle and energy balance and the heating mechanisms in electronegative r.f. discharges. Particular attention is given to the formation of non-Maxwellian electron distributions and their effect on the macroscopic parameters. The research includes theory, particle-in-cell simulation, and experimental investigations. The sheath heating theory and the simulation results developed for electropositive plasmas are used to guide the investigations. The investigation was centered on, but is not limited to, the study of oxygen feedstock gas in capacitively and inductively coupled r.f. discharges.

II Summary of Results

A. Accomplishments in previous contract periods

(a) Plasma Heating. The sheath motion in a capacitively coupled rf

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

discharge is highly nonlinear. We measured the voltage on a floating probe placed in the sheath region, as a function of position and time. A circuit model of the probe-discharge system was used to relate the observed probe voltage to the sheath motion. The results indicate that the primary nonlinear motion is quite similar to the theoretical model. We also have observed oscillations related to the plasma frequency, whose peak harmonic component can be calculated from a simple resonant plasma model. These oscillations can be a useful plasma diagnostic for determining plasma density (publication 3).

We have studied the nonlinear dynamics of stochastic heating arising from the reflection of electrons from moving sheaths as an underlying mechanism for electron power deposition in rf discharges. We showed that the stochastic electron heating leads to a power law electron energy distribution. The heating was determined in both the slow sheath and fast sheath velocity regimes. Commercial low pressure materials processing discharges span both regimes. We numerically investigated the intermediate regime, to obtain a complete picture of the discharge scaling. The scaling and absolute values of the density and absorbed power with voltage was obtained experimentally and compared with the theoretical scaling. Both the scaling and absolute values were similar between theory and experiment (publication 12).

The discharge was simulated using PDP1, a 1-d 3-v, planar, bounded electrostatic PIC code developed by C. K. Birdsall's group in U.C. Berkeley's EECS Department. The electron velocity distribution was found to vary in

time, showing some beamlike behavior. A time average of the distribution can be fitted as a two-temperature Maxwellian or as a power-law distribution. This is in reasonable agreement with experimental measurements and with our previous theoretical study. The scaling of the plasma density and power from the simulation is also in rough agreement with the experimental observations. A paper on these results is in preparation. The combination of analytic, simulation and experimental results has led to good understanding and improved design capability for electropositive rf discharges.

(b) Electron Cyclotron Resonance Discharge. We have investigated the power absorption mechanisms in ECR discharges used for materials processing. This discharges exhibit a sudden transition from a high density regime "high mode" at high power to a low density regime "low mode" at low power. Experiments were performed at 0–700 W in argon at 0.13 mTorr in a cylindrical source chamber propagating TE_{11} mode 2.45 GHz microwave power. We showed that low mode was characterized by a standing wave throughout the plasma chamber and minimal power absorption. High mode exhibited nearly complete power absorption and no standing wave past the ECR zone. A sliding short was used to show that the position of an E field null in the source chamber affected the transition between these two modes for various magnetic field configurations (publication 2).

A formalism was developed for numerically calculating reflection and absorption of the right- and left-hand circularly polarized waves propagating into a cyclotron resonance zone in a medium in which both magnetic field

and plasma density are spatially varying. The absorption and reflections were calculated for various boundary conditions along the propagation path, showing that there is an optimum density for power absorption. The collisionless absorption in the resonance zone, calculated from the single particle heating averaged over the flux, was shown to be consistent with the linear wave absorption at low absorption and collisionality. At high absorption a prescription for bringing the two calculation methods into agreement was developed. The results qualitatively explain the transition from low mode to high mode with increasing power, and show that it is clearly desirable to operate a processing discharge in high mode (publication 4).

(c) Helical Resonator Source. A helical resonator plasma source is a slow wave, high Q structure that may be used for plasma-assisted materials processing at very low pressure ($p \lesssim 1$ mTorr). The resonator consists of a helical coil surrounded by a grounded coaxial cylinder. The source can produce high plasma densities over a wide pressure range, requires no external matching network, and uses no external magnetic field coils. We characterized the helical resonator by measuring the plasma density, resonance frequency, helix voltage, electric field, and Q, for varying pressures and rf powers. We compared these measurements to a model of the resonator operation which predicts the dispersion relation, the field structure, and the scaling of the ohmic and stochastic heating. A self-consistent model was developed for the overall power balance in which we considered the cylindrical geometry and ionization and power loss within the sheath. This model

has been compared with the experimental measurements, giving reasonable agreement (publications 9 and 13).

B. Accomplishments in final contract period

A macroscopic analytic model for a three-component electronegative plasma has been developed. Assuming the negative ions to be in Boltzmann equilibrium, a positive ion ambipolar diffusion equation has been found. The resulting equilibrium equations have been solved in various approximations and compared to a simulation of a parallel-plane r.f. driven oxygen plasma giving reasonable agreement for $p = 50$ mTorr and $n_{e0} = 2.4 \cdot 10^{15}$ m^{-3} (publication 11).

The above comparison was made at an intermediate pressure and electronegativity in which the electropositive edge region can be treated within a constant diffusion model and the negative ion profile in the electronegative region can be approximated by a parabola. We have extended the treatment to other pressure and electronegativity ranges for which other approximations must be made.

For a sufficiently large central ratio, α_0, of negative ions to electrons, the edge electropositive region disappears. The positive-negative ion recombination may dominate the diffusive flow loss, leading to a nonlinear diffusion equation with a solution determined in terms of elliptic integrals. The profile, obtained from this solution has been compared to the parabolic profile approximation, which is good when the flow dominates the recombination. A paper concerned with these results is in progress. Building on the results

of this work, and the simpler approximations used to obtain a *global model* for the equilibrium (publications 6 and 10) an improved global model has been developed and applied to model a capacitively driven plasma etcher. The molecular gases considered consist of either pure chlorine species or a mixture of chlorine and helium species. Analytic scaling laws for the dependence of charged and neutral particle densities, electron temperature, RF voltage and current, sheath width and plasma impedance, on pressure and absorbed RF power, are presented and used to explain the numerical results obtained from the global model. The model results are compared to recent experimental measurements in a chlorine discharge over a range of absorbed power $P_{abs} = 20 - 180$ W at an inlet pressure $p_{in} = 0.4$ Torr and a range of pressure 0.1–1.6 Torr with fixed input power of 100 W. We obtain reasonable agreement for $P_{abs} < 200$ W and for 0.2 Torr $< P_{in} <$ 1 Torr (publication 14).

We have also extended the analysis to lower pressure discharges, for which a variable diffusion model must be employed in the electropositive edge region. We have developed a theoretical description of the edge region, including the variable mobility and the boundary flow conditions. The effect of reaching the local ion sound velocity in the electronegative region has also been considered (publication 15).

III Publications

1. M. A. Lieberman, A. J. Lichtenberg, and S. E. Savas, "Model of Mag-

netically Enhanced, Capacitive RF Discharges," *IEEE Transactions on Plasma Science*, vol. 19, no. 2, pp. 189–196, 1990.

2. D. A. Carl, M. C. Williamson, M. A. Lieberman, and A. J. Lichtenberg, "Axial RF Electric Field Intensity and Ion Density during Low to High Mode Transition in Argon Electron Cyclotron Resonance Discharges," *Journal of Vacuum Science and Technology B*, vol. 9, no. 2, pp. 339–347, 1991.
3. B. P. Wood, M. A. Lieberman, and A. J. Lichtenberg, "Sheath Motion in a Capacitively Coupled Radio Frequency Discharge," *IEEE Transactions on Plasma Science*, vol. 19, no. 4, pp. 619–627, 1991.
4. M. C. Williamson, A. J. Lichtenberg, and M. A. Lieberman, "Self-Consistent Electron Cyclotron Resonance Absorption in a Plasma with Varying Parameters," *Journal of Applied Physics*, vol. 72, no. 9, pp. 3924–3933, 1992.
5. M. A. Lieberman and R. A. Gottscho, "Design of High Density Plasma Sources for Materials Processing," in *Physics of Thin Films*, ed. by M. Francombe and J. Vossen, Academic Press, New York, 1993, 119 pages.
6. C. Lee, D. B. Graves, M. A. Lieberman, and D. W. Hess, "Global Model of Plasma Chemistry in a High Density-Oxygen Discharge," *J. Electrochemical Society*, vol. 141, pp. 1546–1555 (1994).
7. P. N. Wainman, M. A. Lieberman, A. J. Lichtenberg, R. A. Stewart, and C. Lee, "Characterization at Different Aspect Ratios (Radius/Length) of a Radio Frequency Inductively Coupled Discharge," *J. Vac. Sci.*

Technol., **13,** 2464 (1995).

8. M. A. Lieberman and A. J. Lichtenberg, *Principles of Plasma Discharges and Materials Processing*, John Wiley and Sons, New York, 1994, 572 pages.
9. K. Niazi, A. J. Lichtenberg, M. A. Lieberman, and D. L. Flamm, "Operation of a Helical Resonator Source," *Plasma Sources Sci. Technol.*, vol. 3, pp. 482–495 (1994).
10. C. Lee and M. A. Lieberman, "Global Model of Ar, O_2, Cl_2 and Ar/O_2 High Density Plasma/Discharges," *Journal of Vacuum Science and Technology A*, **13,** 368 (1994) 21 pages.
11. A. J. Lichtenberg, V. Vahedi, M. A. Lieberman, and T. Rognlien, "Modeling Electronegative Plasma Discharges," *J. Appl. Phys.*, **75,** 2339 (1994).
12. B. Wood, M. A. Lieberman, and A. J. Lichtenberg, "Stochastic Electron Heating in a Capacitive R.F. Discharge," *IEEE Trans. Plasma Sci.*, **23,** 89 (1995).
13. K. Niazi, A. J. Lichtenberg, and M. A. Lieberman, "The Dispersion and Matching Characteristics of the Helical Resonator Plasma Source," *IEEE Trans. on Plasma Sci.*, **23,** 833 (1995).
14. Y. T. Lee, M. A. Lieberman, A. J. Lichtenberg, F. Bose, H. Baltes, and R. Patrick, "Global Model for High Pressure Electronegative Radio-Frequency Discharge," *J. Vac. Sci. and Technol.*, to appear (1996).
15. I. Kouznetsov, A. J. Lichtenberg, and M. A. Lieberman, "Modeling an

Electronegative Discharge at Low Pressure," *Plasma Sources Sci. and Technol.*, to appear (1996).

IV Graduate Students

Blake Wood

Chris Lee

Mike Williamson

Peter Wainman

Igor Kouznetsov

Zouding Wang

Y. T. Lee (visitor, not supported)