	こんどうしゅうじ
氏名	近藤修司
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指導教官	東北大学教授　南部　健一
論文審査委員	主査　東北大学教授　南部　健一　　東北大学教授　西山　秀哉 東北大学教授　江刺　正喜　　東北大学教授　佐藤　徳芳

論文内容要旨

Chapter 1 Introduction

The application of plasma technology to semiconductor fabrication, i.e., plasma processing is one of the great successes of sciences. Sputtering using dc- or rf-driven magnetron discharge is one of the most commonly used techniques for fabrication of thin films. Currently, the size of silicon wafer or liquid crystal display is increasing year by year. Consequently, development of sputtering apparatuses based on experimentally obtained design parameters is becoming exorbitantly expensive. The method of accurately simulating magnetron discharge structures is expected to reduce the cost of research and development. Therefore considerable efforts have been made to model magnetron discharges using the various models. Typical pressure in planar magnetron discharges is less than 5 mTorr. Hence, to obtain a detailed description of the magnetron discharge we should use particle models. Objective of this thesis is to clarify the structure of dc and rf magnetron discharges by use of the particle-in-cell/Monte Carlo (PIC/MC) method. Particularly, modeling of rf magnetron discharges, which is really a challenging problem because of its intensive computational task, is successfully carried out for the first time.

Chapter 2 Analysis of Electron Swarm in Argon by the Test Particle Monte Carlo Method

The electron–atom collision should be treated as accurately as possible to perform reliable discharge analysis. In the analysis of magnetron discharges in the following chapters, argon gas is assumed as working gas of discharge. Therefore, in chapter 2 the analysis of the electron swarm in argon was performed to verify the validity of the collision model and the cross sections for electron–argon-atom collisions used in the present discharge analysis. The swarm simulation was performed by use of the test particle Monte Carlo method to obtain electron swarm parameters such as drift velocity, diffusion coefficient, and ionization coefficient. These swarm parameters were compared with experimental and calculated data of other researchers. The results are summarized as follows.

1. Agreement of the obtained swarm parameters with other experimental and calculated data was found to be very well; even the snapping of the swarm parameters at a certain value of reduced electric field was reproduced.
2. Also it was found that in high energy region the swarm parameters are strongly affected by the forward scattering of electrons, *e.g.*, the snapping of the drift velocity results from increasing forward scattering.

3. Therefore, the application of the present collision model to discharge analysis of argon plasma is recommendable and hence, reliable discharge simulation can be expected.

Chapter 3 Three-Dimensional Analysis of DC Planar Magnetron Discharge by the Particle-in-Cell/Monte Carlo Method

A particle-in-cell/Monte Carlo simulation method applicable to the analysis of three-dimensional dc planar magnetron discharges was proposed. Using this method, the structure of dc magnetron discharge with an axisymmetrical magnetic field was clarified; the effects of magnetization M of the magnet and the secondary electron emission coefficient γ on the discharge structure were examined. The axisymmetrical magnetic field is formed by the two concentric cylindrical magnets on the back of the target. The centers of N and S poles are at $r = 25$ and 50 mm, respectively, where r is the radial distance measured from the axis of the magnets. The results are summarized as follows.

1. The spatial distributions of the electric field, electron and ion densities, and charge density clearly show the formation of an axisymmetrical sheath near the cathode. The thickness of the sheath varies in the radial direction and the sheath is thinnest at the mid-point of N and S poles. The weak sheath is formed in the vicinity of the anode. The overall features of electric field and electron density near the mid-point of the two poles ($r = 41$ mm) are analogous to those for unmagnetized discharge.

2. The thickness of the cathode sheath at the mid-point decreases with increasing magnetization M or emission coefficient γ. As M increases, electrons are trapped in the region nearer to the cathode. This trapping results in shifting the location of the peak ionization rate and hence the peak electron density toward the cathode. Since plasma is apt to keep charge neutrality, the location of the peak ion density is also shifted toward the cathode. Consequently, as M increases, the location of the peak charge density is shifted toward the cathode and hence decrease in sheath thickness occurs. The simulated results clearly support this physical image. The sheath thickness d_s calculated follows well Lan Gu and Lieberman's similarity law ($d_s \propto V^{7/8}/I^{1/2}B^{1/4}$). The effect of γ on the magnetron discharge is qualitatively analogous to the dc glow discharge.

3. The effect of M on the electron density profile has shown a qualitative agreement with that on the measured emission profile although the latter was obtained under a constant current, not constant voltage as in the present work.

4. The ion flux onto the cathode is uniform in the circumferential direction; no instability appeared for the four cases studied. Also the spatial distributions of mean electron and ion energy are examined. The distribution of the electron energy reflects the complicated $\boldsymbol{E} \times \boldsymbol{B}$ motion. The ion energy takes the maximum on the cathode. The maximum ion energy is about 60% of the cathode potential. The peak positions of ionization rate, excitation rate, and electron density are different; the peak of ionization rate is closest to the cathode and the peak electron density is farthest from the cathode.

Chapter 4 Chaotic Dynamics of Three-Dimensional DC Magnetron Discharge

Magnetron sputtering apparatuses with large racetrack-like magnets are often used for thin film deposition on liquid crystal displays. In this chapter a full three-dimensional PIC/MC simulation of dc magnetron discharge with racetrack-type magnets have been performed to examine low-frequency turbulence in the discharge. The results are summarized as follows.

1. Some kind of low-frequency wave appeared in plasma under certain discharge conditions of low applied voltage or weak magnetic field strength.

2. The power spectra of the electron density fluctuations were found to be continuous, which means a chaotic change.
3. Such a low-frequency plasma turbulence were experimentally observed in dc magnetron discharges by others. Therefore, PIC/MC simulations by use of a supercomputer such as the one reported in this work may make it possible to open the veil of complicated phenomena caused by a collective motion of plasma.

Chapter 5 Axisymmetrical Analysis of DC Planar Magnetron Discharge by the Particle-in-Cell/Monte Carlo Method

The analysis in chapter 3 showed that the discharge is free of azimuthal instabilities and truly axisymmetrical for the four discharge conditions studied. Therefore in the present chapter a fast Poisson solver for axisymmetrical electric fields was developed to analyze axisymmetrical discharges. By use of the solver an axisymmetrical PIC/MC analysis of dc magnetron discharge was performed for the same magnetron sputtering apparatus as that treated in chapter 3, assuming that the electric field is axisymmetrical. The effects of the applied voltage and magnetization on the plasma structure and current–voltage characteristics have been clarified. The results are summarized as follows.

1. The cathode sheath thickness d_s increases as the applied voltage V_c increases, and d_s decreases with increasing the magnetization M of the magnets. This can be explained from the effects of V_c and M on the location of collisionless cycloidal $\boldsymbol{E} \times \boldsymbol{B}$ motion of electrons near the cathode; the location is shifted toward the plasma bulk with increasing V_c and toward the cathode with increasing M. The effect of V_c on the sheath thickness is similar to that of unmagnetized discharge.
2. Differently from many experimental results, the simulated discharge showed a negative current–voltage characteristics; the current I peaks at a moderate voltage 400 V and then decreases with a further increase of the voltage. The result can be explained based on Lan Gu and Lieberman's similarity law $I \propto V_c^{7/4}/d_s^2 M^{1/2}$ for the magnetron discharge with circular magnetic field lines, where d_s is the sheath thickness. At voltage less than 400 V, the effect of V_c on the sheath thickness is small and hence the current increases with voltage. As the applied voltage V_c increases further more, the sheath thickness d_s increases; increase of d_s causes reduction of the ion current due to the energy loss of Ar^+ by increased elastic and charge exchange collisions. That is, there is an optimum voltage for the magnetron discharges. For the optimum voltage (400 V), the ion current is maximal and plasma density is highest. This is due to the best coupling of electric and magnetic fields. For voltage higher than 400 V, ionization by secondary electrons occurs less efficiently because of the reduction of ionization cross section. It is concluded that the essential characteristics of the present dc magnetron discharge can be explained based on Lan Gu and Lieberman's similarity law. In general, the ion current increases with decreasing the ratio of ion energy loss to applied voltage.

Chapter 6 Axisymmetrical Analysis of RF Planar Magnetron Discharge by the Particle-in-Cell/Monte Carlo Method

An axisymmetrical particle-in-cell/Monte Carlo analysis of rf planar magnetron discharges was performed. The simulation procedure used here is the same as that used in chapter 5, except for an addition of the treatment of dc self-bias which appears on a powered electrode in rf-driven discharges. The discharge structure of argon at frequency of 13.56 MHz and voltage amplitude of 200 V was clarified for four discharge conditions. That is, various plasma parameters such as electric field, plasma density, discharge current, electron power deposition, and electron and ion energy were examined by varying the magnetic field strength and the secondary electron emission coefficient. The results are summarized as follows.

1. The spatial and temporal distributions of the electric field, electron and ion densities, and charge density clearly show the formation of an axisymmetrical sheaths near the powered and grounded electrodes. The time-modulation of the sheaths is much smaller than the case of rf discharges with no magnetic field.
2. Even in the bulk region of the plasma, the electric field is noticeably time-modulated, which never occurs in usual rf argon discharges without magnetic field.
3. The effects of magnetization M of the magnet and secondary electron emission coefficient γ on the electric field, plasma density, and charge density are similar to those in dc magnetron discharges, *e.g.*, the sheath thickness decreases with increasing M or γ.
4. The dc self-bias, V_{dc}, that appears on the target (powered electrode) due to the asymmetry of the discharge caused by the applied dc magnetic field is slightly positive for $M = 0.25\,\mathrm{T}$ and it decreases and becomes negative with a further increase of M.
5. The effect of M on V_{dc} is contrary to other's experiments of magnetron discharges performed under the condition that the magnetic field is parallel to the target (powered electrode). The different effect of M on V_{dc} between the present results and the experiments can be explained from a different mechanism of electron transport to the target; in the present model electrons moving along circular magnetic field lines can reach the target through the regions where the magnetic field lines are nearly perpendicular to the target, whereas electrons have to traverse the magnetic field lines to reach the target in the experiments referred.
6. The spatial and temporal distributions of the total discharge current show that the discharge is capacitive even in the rf magnetron discharge. As M or γ increases, the phase difference between the current and the applied voltage becomes slightly smaller in agreement with experimental results.
7. The larger ion conduction current flows to the powered electrode (target) than to the grounded electrode due to the asymmetry of the discharge. This is due to localization of ion density near the target.
8. The displacement current is large in the bulk region owing to a large time-modulation of bulk electric field.
9. Electron power deposition occurs substantially in the bulk region due to an appreciable electric field in the bulk. The total power deposition in the discharge space is maximal for a medium magnetic field at a fixed γ.
10. The mean electron energy in the bulk region is 6–10 eV. The mean ion energy is high near the two electrodes because of ion acceleration by the sheath electric fields.
11. Inelastic collisions of electrons are the main loss mechanism of the input power. That is, the total power deposition is largest for such a discharge condition that the overall inelastic collision rate is maximal.

Chapter 7 Conclusions

First, electron–argon-atom collision model was verified by the test particle Monte Carlo method in advance of its use in the discharge analysis. Next, the particle-in-cell/Monte Carlo (PIC/MC) simulations were performed for modeling dc- and rf- planar magnetron discharges with axisymmetrical or three-dimensional magnetic field to clarify the discharge structures and the correlation between system parameters and plasma parameters. In a new future, detailed simulations of discharges such as the ones reported here will become the only tool to unveil complicated phenomena in other types of magnetized or non-magnetized plasmas.

審査結果の要旨

直流および高周波マグネトロンプラズマを用いた高速低温スパッタは，金属や絶縁体の薄膜作成に幅広く使われている．そのため数値シミュレーションによるその詳細な放電構造の解明が半導体産業等から強く求められており，実験・試作を伴わないシミュレーションのみによるスパッタ装置の設計・開発が期待されている．本論文は，直流および高周波マグネトロンプラズマ解析のための粒子シミュレーション法を開発し，これを用いてプラズマ構造の詳細を解明したもので，全編7章よりなる．

第1章は緒論であり，本研究の背景および目的を述べている．

第2章では，第3章以降のプラズマ解析で用いる電子–アルゴン原子衝突モデルの妥当性の検証を，テスト粒子モンテカルロ法によるスオームシミュレーションにより行っている．求められた電子スオームパラメータは既存の実験結果と良く一致しており，信頼性の高いプラズマ解析が可能であることを示している．また前方散乱を取り込んだ微分断面積がスオームパラメータに大きな影響を持つこと明らかにしている．

第3章では，軸対称磁場を持つ平行平板直流マグネトロン放電の3次元解析を行い，電場，プラズマ密度，イオンフラックス，電子エネルギー，イオンエネルギー等の空間分布に対する，磁場強度と二次電子放出係数の影響を明らかにしている．またプラズマは解析を行った放電条件では周方向に不安定性を示さないことも明らかにしている．得られた結果は実験結果とも一致している．これは，マグネトロンスパッタリング装置の計算機支援設計の実現にとって重要な成果であると言える．

第4章では，レーストラック型の永久磁石を持つ直流マグネトロン放電について3次元解析を行い，磁場強度または印加電圧が小さい時には，周方向に低周波不安定性が発生することを明らかにしている．電子数密度の時系列のパワースペクトルは連続となり，カオス的な変化が起こっていることを示しており興味深い．このような低周波不安定性は実験によっても報告されており，精密なPIC/MCシミュレーションがプラズマの複雑な協同現象の解明にも有用であることを示したことは重要である．

第5章では，まずフーリエ正弦変換を用いた軸対称電場の高速解法を開発している．この解法は計算時間の大幅な減少を可能にしており，PIC/MC解析の計算負荷を現実的なものにすることに大きく貢献している．次に開発した解法を用いて直流マグネトロン放電の軸対称解析を行い，放電構造に対する印加電圧，磁場の影響を調べている．放電構造の印加電圧に対する特性はGuとLiebermanの法則に良く一致することを見出しており，有用な知見と言える．

第6章では，第5章で開発した軸対称PIC/MC法をもとに高周波マグネトロン放電の解析を行い，その時空間構造に対する磁場と二次電子放出係数の影響を示している．まず，パワー側電極における自己バイアス電位が，磁場によるプラズマの非対称によって発生することを解明している．また，電場はバルク中でも強い時間変調を受けること，およびパワー側電極近傍では磁場による電子運動の拘束によりほとんど時間変調されないことを解明した．これらの結果は高周波マグネトロン放電の構造が磁場のない高周波放電の構造とは本質的に異なることを見出したものであり，重要な成果である．

第7章は結論である．

以上要するに本論文は，直流および高周波マグネトロンプラズマの時空間構造を解明し，種々のプラズマパラメータと外部パラメータの関係を明らかにしたもので，プラズマ工学・電子工学の発展に寄与するところが少なくない．

よって，本論文は博士(工学)の学位論文として合格と認める．