Realization of novel plasma performances based on systematic understanding of plasma behaviour using laser diagnostics

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Abstract

Laser diagnostics have been extensively used to understand plasma behaiviour under different discharge conditions. Measurements were performance for (i) electric field, (ii) electron temperature and density, and (iii) reaction products due to chemical reactions by electron impacts. The knowledge thus gained has been extensively used to realize novel plasma performances, such as epitaxial thin film depositions using plasma sputtering, performance improvements of discharge-pumped excimer lasers, and developments of environmental equipment.

1. Introduction

In plasma applications, controllable discharge parameters, such as electrode configuration, input power, gas composition and flow rate, and pressure, are changed over a wide range in order to find suitable conditions for various applications. However, this trial and error approach is becoming more and more difficult because requirements for those applications, such as finer etching, better quality thin films, higher performance light emission, better laser performance, and better plasma confinement for thermonuclear fusion are becoming more and more stringent. This is a strong motivation to gain better understanding and control of plasma parameters.

In a very simplistic expression, the input power in plasma discharges is first (1) fed into the working gas as an electric field, (2) which then accelerates electrons to be heated, (3) the electrons subsequently collide with constituent gases to form various radicals, excited atoms and molecules, and/or ions, and (4) the radicals/atoms/ molecules/ions which are produced execute useful actions, such as those listed above.

We can summarize the aim of the laser-aided plasma diagnostics (LAPD) as the measurement of important plasma parameters in the above three processes, namely, (1) electric fields, (2) electron density and temperature, and (3) neutral and ionic particles, using laser diagnostics, so as to find optimum conditions for (4). This situation is summarized in Fig. 1.

Until now, LAPD has been developed on rather individual bases, namely to try to meet various measurement requirements by applying/improving well-established methods or by developing new methods /techniques. Bearing these in mind, we have devoted, for the last 30 years, a lot of effort to develop LAPD from the viewpoints (1) ~ (3) above, so as to gain an unified understanding of various discharge processes. Based on such an understanding, we have recently succeeded in realizing useful plasma actions, which would have been otherwise impossible.

In this article, one example of such novel plasma actions, namely an epitaxial growth of single crystal Si-thin films using plasma sputtering, is described based on the successful developments of various LAPD methods. In Section 2, the background of the present epitaxial growth is described. In Section 3, researches leading to the present project are described. Experimental procedures and results are described in Section 4. Section 5 contains discussion about this research, while Section 6 contains concluding remarks.

2. The implication of the present Si epitaxial growth

Si epitaxy is used in integrated circuit technology to create single-crystal Si layers on top of Si substrates, because the resistivities and dopant types of the layers can be adjusted independently of those of the substrates. We have recently succeeded in producing Si epitaxial deposition in a plasma system with a low substrate temperature of 400℃ and conventional base vacuum of 5×10^{-7} Torr [1, 2]. Our main concepts include the following three points: (1) to use a plasma stream having a high flux and low ion energy to bombard the substrate surface for cleaning; (2) to provide sufficient activation energy to surface adatoms on the substrate during the deposition process, in place of the conventional high temperature heating, so that the adatoms can migrate on the surface and find themselves at normal lattice sites to form a single crystal; and (3) to deposit Si at a high flux with the monolayer formation time in the range of 1-2 s, which is much shorter than the impurity monolayer formation time of 23 s at the conventional base vacuum of 5×10^{-7} Torr.

This deposition of high quality thin films has become possible by a collaboration of plasma and electronics researchers. In particular, we emphasize that LAPD in the plasma research group has played an important part.

3. Researches to the present project

During the last 25 years, we have systematically studied various sputtering processes, motivated initially by the necessity of erosion control for the realization of high temperature plasmas for thermonuclear fusion research. In order to measure densities and velocity distribution functions of various impurities that are released from walls into plasmas, with sufficient spatial and temporal resolution, we embarked on developing the technique of laser-induced fluorescence (LIF). After a preparatory phase [3], the technique has been applied to two directions of projects; firstly to the study of particle behavior in high-temperature plasmas and secondly, to the study of fundamental sputtering processes. The former was first studied in Heliotron E and subsequently in JT-60U at Japan Atomic Energy Research Institute (JAERI) [4]. The latter has developed into the study described in this article.

The first project of the fundamental sputtering process research was the study of the energy dependence of angular distribution of sputtered atoms from a target by ion beam bombardment [5], which evolved into a collaboration with a theoretical group [6]. Then, after development of a RAFS (rapid frequency scan) dye laser [7] by collaboration with a laser group, this research evolved into a very successful work of transient change of a sputtering process of gradual removal of oxidized layers into a pure metal sputtering [8] .

During the course of these studies, it was realized that plasma sputtering was being used as a useful means of materials deposition, but that the governing processes had not been well understood. Following up the understanding of the particle ejection from solid surfaces by ion bombardment described above [5, 6, 8], sputtered particle transport in plasmas was studied, and a clear correlation of particle thermalization with the gas pressure was formulated based on the ratio of transported distance of ejected particles against mean free path lengths [9].

At about the same time that we initiated the above plasma sputtering research, a new class of "high density" glow discharge plasmas, such as ECR (electron cyclotron resonance) and ICP (induction coupled plasma), were starting to be used as plasma sources for materials processing. Here again, the mechanisms of plasma formation and maintenance were not well understood. We embarked on trying to understand first the electron behaviour [10, 11] and then ion behaviour [12] in ECR plasmas.

When a new research center was created in Kyushu University in 1994, called the "Advanced Science and Technology Center for Cooperative Research (ASTEC)", a collaborative research between the plasma group, which led the above-mentioned projects, and electronic and chemical researchers was begun. As a common tool for the joint project, an ECR sputtering apparatus was selected. After a preparatory phase, in which several high quality oxide and oxynitride films were successfully deposited [13, 14] and their formation mechanisms were studied [15], an ambitious project was initiated in 1997, leading to the present success.

4. Experimental procedures and results

Details of experimental apparatus, procedures and results are described elsewhere [1, 2], and relevant features leading to the plasma physics discussions in Section 5 are shown here.

A sputtering-type electron cyclotron resonance (ECR) plasma was used for epitaxial deposition of Si on Si substrates. The apparatus is schematically shown in Fig. 2. Microwaves at a frequency of 2.45 GHz were introduced through rectangular waveguides. Two magnet coils were set around the plasma chamber to supply the ECR magnetic field in the plasma chamber, which diverged from the microwave composer to the deposition chamber for plasma extraction. The magnetic flux density of 875 G for the ECR condition was at around the chamber center.

The target was *n*-type Si. The substrate was a *p*-type (100) Si single crystal, which was first chemically cleaned and then immediately loaded into the vacuum chamber, which was subsequently pumped down to a base pressure of less than 5×10^{-7} Torr. The substrate was gradually heated up to 400℃ for 1 h, then the plasma was operated in order to clean the surfaces of the target and substrate. The deposition was then started by applying RF power to the target.

The structure of the deposited films was evaluated using spectroscopic ellipsometry and electron backscattering diffraction (EBSD) techniques.

The effect of the gas pressure during deposition on the film quality was investigated in the range of 0.5-2.5 mTorr. Figure 3 shows the results for the samples deposited at gas pressure of 0.7, 1.7 and 2.5 mTorr for the same in situ cleaning conditions listed in Table 1. The k spectrum of the sample deposited at the gas pressure of 0.7 mTorr was different from that of the Si substrate, while for 1.7 and 2.5 mTorr, the spectra were very close to that of the Si substrate. EBSD results for the latter samples showed a very clear Kikuchi line pattern, indicating that these were single crystal thin films.

Figure 4 demonstrates the effect of deposition rate, which was changed by P_{rf} to the target, on the epitaxial deposition, where parameters listed in Table 1 were used for the cleaning and deposition. When the deposition rate was 0.03 Å/s, corresponding to P_{rf}= 62 W, the deposited films was obviously amorphous. The k spectrum of the film deposited at 0.2 Å/s (P_{rf}=100 W) was somewhat different from that of the Si substrate. However, the films deposited at rates of more than 0.6 Å/s (P_{rf}=200 W) showed the same characteristics, which were close to that of the crystal Si.

5. Discussion

The ion bombardment energy of ions striking the substrate is an important parameter in the ECR plasma system. It is evident from the foregoing observations that there exists a clear difference in the film quality for deposition at gas pressures above and below 1 mTorr. This is due to the fact that the increase of gas pressure reduces the ion energy, due to collisions and/or distributed ionization near to the substrates. Recent preliminary results of substrate biasing experiments clearly indicate that ion energy had a profound influence on the film quality. When the bombardment ion energy is excessive, some damages on the substrate/film must be caused, resulting in amorphous formation.

In order to obtain epitaxial growth, a sufficient amount of surface activation energy must be provided so that surface adatoms can migrate on the surface and locate themselves at normal lattice sites. This energy can be supplied by heating the substrate (over 1000℃) in the conventional method, such as CVD. However, Ar ion bombardment is useful for supplementing insufficient thermal energy, leading to epitaxial growth at low temperature. We believe that the sum of the Si deposition energy and flux of Ar ions is the main factor which compensates for the reduction in the substrate temperature. Thus, the fact that epitaxial growth could not be achieved below 350℃ may be explained by an insufficient compensation.

Another key factor is the competition between fluxes of Si atoms and residual gas impurities onto the substrate surface. Figure 5 shows that the flux of Si atoms has a great influence on the film structures. A low P_{rf} of 62 W causes a small flux of Si atoms (1.3×10^{17} $m^{-2}s^{-1}$), which corresponds to Si monolayer formation time of 41 s. In contrast, the impurity monolayer formation time is about 23 s, which is shorter than Si monolayer formation time, suggesting that an impurity monolayer was formed faster than the Si monolayer. Therefore, it is well understood that the deposited films were amorphous. When P_{rf} was increased from 200 to 500 W, the Si monolayer formation time is in the range of 1-2 s, implying that the Si monolayer grew on the substrate much more quickly than an impurity monolayer. Thus, an epitaxial Si film was obtained.

6. Conclusions

Si epitaxial film was deposited for the first time at the conventional base vacuum of 5×10^{-7} Torr and low substrate temperature of 400℃. The two key issues in achieving this were (i) control of the ion energy upon the substrate/film and (ii) use of a high flux of Si atoms supplied by a sputtering-type ECR plasma. Another important factor in this success is the collaboration of researchers in completely different backgrounds and disciplines, which have been required at every step of this kind of interdisciplinary work.

References

[1] J. Gao, H. Nakashima, N. Sakai, D. Gao, J. Wang, K. Furukawa and K. Muraoka, *Jpn. J. Appl. Phys.* **38** (1999) L220.

[2] J. Gao, J. Wang, N. Sakai, K. Iwanaga, K. Muraoka, H. Nakashima, D. Gao and K. Furukawa, *Jpn. J. Appl. Phys.* (submitted).

[3] M. Hamamoto, M. Maeda, K. Muraoka and M. Akazaki, *Jpn. J. Appl. Phys.* **20** (1981) 1709.

[4] K. H. Takenaga, K. Shimizu, N. Asakura, S. Tsuji-Iio,M. Shimada, M. Kikuchi, K. Uchino and K. Muraoka, *Nucl. Fusion.* **35** (1995) 853.

[5] Y. Matsuda, Y. Yamamura, Y. Ueda, K. Uchino, K. Muraoka, M. Maeda and M. Akazaki, *Jpn. J. Appl. Phys.* **25** (1986) 8.

[6] Y. Yamamura and K. Muraoka, *Nucl. Instr. Meth. Phys. Res.* **B42** (1989) 175.

[7] C. Honda, K. Nishimura, M. Maeda, K. Muraoka and M. Akazaki, *J. Phys. D: Appl. Phys.* **16** (1983) 1943.

[8] Y. Matsuda, C. Honda, S. Matsubaguchi, T. Moroishi, K. Muraoka, M. Maeda and M. Akazaki, *Jpn. J. Appl. Phys.* **25** (1986) L182.

[9] W. Z. Park, T. Eguchi, C. Honda, K. Muraoka, Y. Yamagata, B. W. James, M. Maeda and M. Akazaki, *Appl. Phys. Lett.* **58** (1991) 2564.

[10] M. Bowden, T. Okada, F. Kimura, H. Muta, K. Uchino, K. Muraoka, T. Sakoda, M. Maeda, Y. Manabe, M. Kitagawa and T. Kimura, *J. Appl. Phys.* **73** (1993) 2732.

[11] T. Hori, M. Kogano, M. D. Bowden, K. Uchino and K. Muraoka, *J. Appl. Phys.* **83** (1998) 1909.

[12] W. Cronrath, N. Mayumi, M. D. Bowden, K. Uchino, K. Muraoka and M. Yoshida, *J. Appl. Phys.* **82** (1997) 1036.

[13] H. Nakashima, K. Furukawa, Y. C. Liu, D. W. Gao, Y. Kashiwazaki, K. Muraoka, K. Shibata and T. Tsurushima, *J. Vac. Sci. Tech.* **A15** (1997) 1951.

[14] D. W. Gao, Y. Kashiwazaki, K. Muraoka, H. Nakashima, K. Furukawa, Y. C. Liu, K. Shibata and T. Tsurushima, *J. Appl. Phys.* **82** (1997) 5680.

[15] K. Furukawa, Y. Liu, H. Nakashima, D. Gao, Y. Kashiwazaki, K. Uchino, K. Muraoka and H. Tsuzuki, *J. Appl. Phys.* **84** (1998) 4579.

Table 1: Surface cleaning and deposition parameters.

parameter	in situ cleaning	deposition
Substrate temperature (℃)	400	400
Processing time (min)	3	60
Microwave power (W)	500	500
rf-power (W)	----	500
Ar gas flow rate (sccm)	25	25
Ar gas pressure (mTorr)	1.5	1.5

Note: The same in situ cleaning conditions were used for selecting suitable deposition parameters. Similarly, the same deposition conditions were used for selecting cleaning parameters.

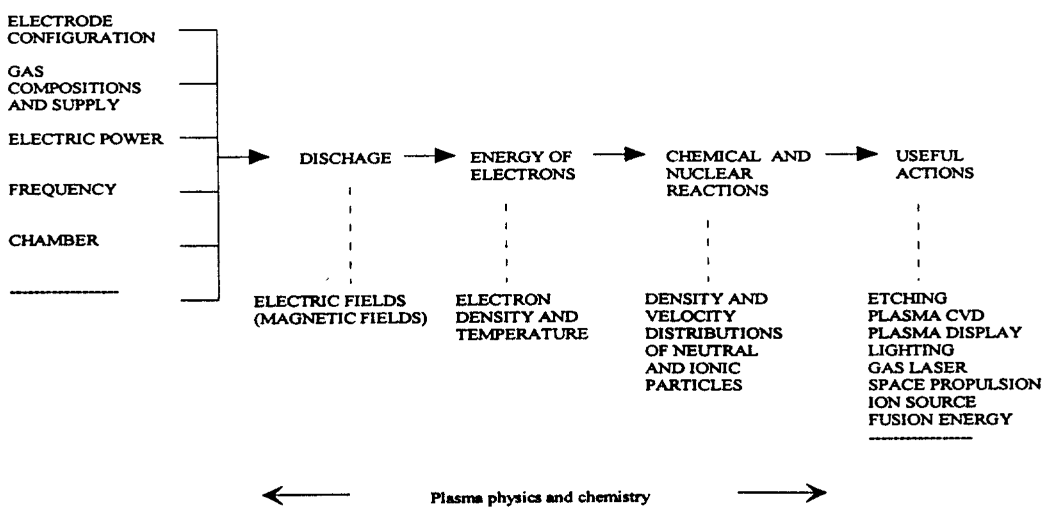

Fig. 1 The role of plasma physics and chemistry

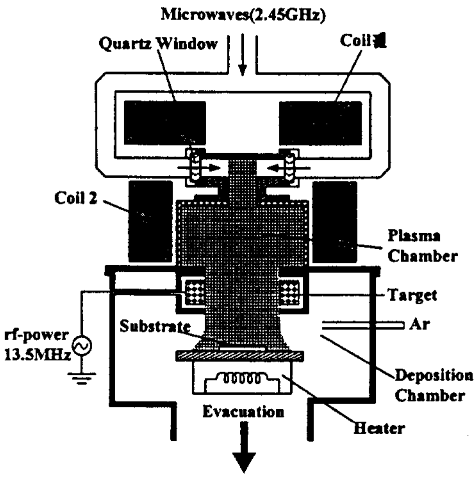

FIG. 2: Schematic diagram of a sputtering-type ECR plasma system.

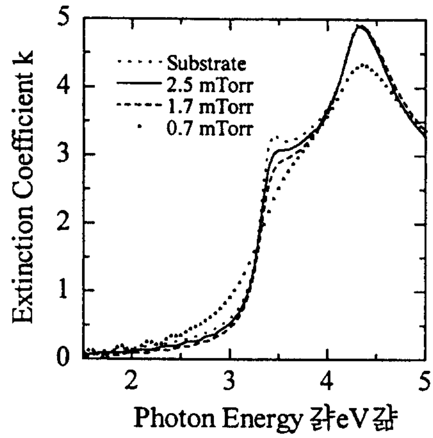

FIG. 3: Dependence of the Ar gas pressure during the deposition on the film quality. The in situ cleaning was carried out under the same conditions as listed in Table 1.

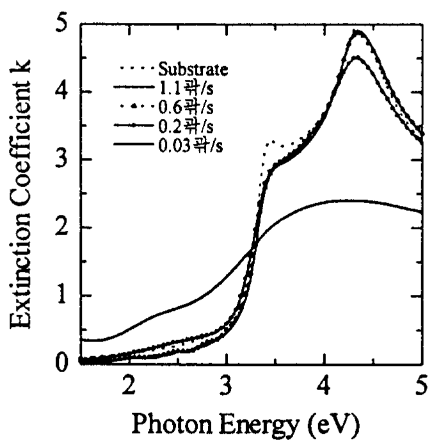

FIG. 4: Effects of the deposition rate on the film quality. The Si films were prepared under the same in situ cleaning and deposition conditions as listed in Table 1.

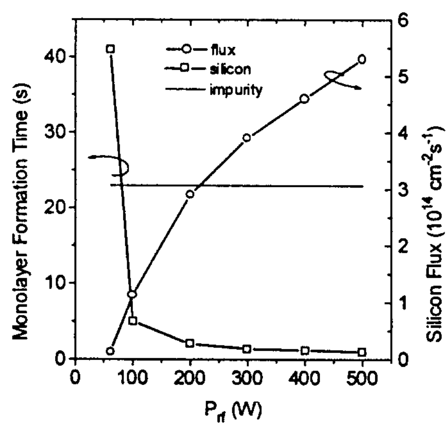

FIG. 5: Flux of Si atoms near the substrate surface and Si monolayer formation time as a function of P_{rf}. As shown by the dotted line, the impurity monolayer formation time is 23 s.