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Plasma chemistry fluctuations in a reactive arc plasma in the presence of magnetic fields

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The effect of a magnetic field on the plasma chemistry and pulse-to-pulse fluctuations of cathodic arc ion charge state distributions in a reactive environment were investigated. The plasma composition was measured by time-of-flight charge-to-mass spectrometry. The fluctuation of the concentrations of Al^+, Al^{2+}, and Al^{3+} was found to increase with an increasing magnetic field strength. We suggest that this is caused by magnetic field dependent fluctuations of the energy input into cathode spots as seen through fluctuations of the cathode potential. These results are qualitatively consistent with the model of partial local Saha equilibrium and are of fundamental importance for the evolution of the structure of films deposited by reactive cathodic arc deposition. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1483128]

Vacuum arc plasma is produced in micrometer-sized cathode spots of very high current, power, and plasma density ($\sim 10^{12}$ A/m^2, $\sim 10^{13}$ W/m^2, and $\sim 10^{26}$ m^{-3}, respectively).[1] The high power density is associated with explosive phase transitions, resulting in a plasma that is highly ionized and which can be used for thin film synthesis, ion implantation, and ion injection into accelerators. The individual ion charge states (Q_i) are of importance in plasma processing because the kinetic energy gain (ΔE_i) across the potential difference between plasma and substrate (ΔU) is given by $\Delta E_i = Q_i\ \Delta U$. It is well known that the kinetic energy, and therefore the charge states, affect the microstructure evolution and hence the film properties.[2]

Experimental as well as theoretical data describing the plasma composition and the resulting average ion charge states of vacuum arc plasmas are available for more than 50 cathode materials.[3] The effect of both the reactive gas pressure as well as the presence of a magnetic field on the pulse-to-pulse fluctuations of the ion species distribution in the plasma have not been investigated, despite the fact that thin film compound synthesis by cathodic arc is usually achieved in the presence of reactive gas as well as magnetic fields.

We used pulsed vacuum arcs to study the plasma chemistry and ion charge state fluctuations in an oxygen background with a varying magnetic field. We found that increasing the magnetic field strength results in an extensive increase in plasma chemistry fluctuations. This may have important consequences for the microstructure evolution during thin film growth.

The analysis of the plasma composition was carried out using a time-of-flight (TOF) charge-to-mass spectrometer at Berkeley Lab, described in detail in Ref. 4. This setup has been used in the past to determine the plasma chemistry of reactive arc plasma in the presence of magnetic fields,[5] however, without investigating the statistics of the large fluctuations inherent to cathodic arc plasmas. Ions are extracted from the plasma source, forming a beam. A set of annular electrostatic TOF gates is used to select a 200 ns sample of the beam, which is detected by a magnetically suppressed Faraday cup located 1.03 m from the TOF gate. The plasma composition can be calculated from the measured ion current–time dependence.

The plasma was generated from an aluminum cathode. The arcs were triggered by a high voltage flashover across a ceramic tube between a trigger electrode and the cathode. The arc current was 400 A and the arc pulse duration was 250 μs at a repetition rate of 1 Hz. All data were taken at 150 μs after the arc was ignited at an oxygen partial pressure of 9×10^{-2} Pa. Prior to introduction of the oxygen the system base pressure was about 9×10^{-5} Pa. A magnetic field could be induced in the immediate vicinity of the cathode by discharging a 250 μF capacitor bank via a high-current thyristor switch and solenoid coil. The coil was 30 mm in length and 20 mm in diameter. The magnetic field at the cathode surface was up to 0.28 T with coil or 0 T when the coil was not used. From 100 individual time-of-flight spectra the ion concentrations for Al^+, Al^{2+}, and Al^{3+} ions were calculated. From the statistical analysis of these data one can learn about plasma chemistry fluctuations with a resolution of 200 ns. The relative standard deviation (S_{rel}) of the individual aluminum ions is shown versus the magnetic field strength in Fig. 1. The magnetic field strength strongly affects the pulse-to-pulse fluctuations (S_{rel}): Without magnetic field, S_{rel} for Al^+, Al^{2+}, and Al^{3+} equals 4%, 7%, and 26%, respectively. As the magnetic field is increased up to a maximum of $B = 0.28$ T, S_{rel} increases drastically to values of up to 46%, 66%, and 91%, respectively.

We have also measured the arc burning voltage, i.e., the

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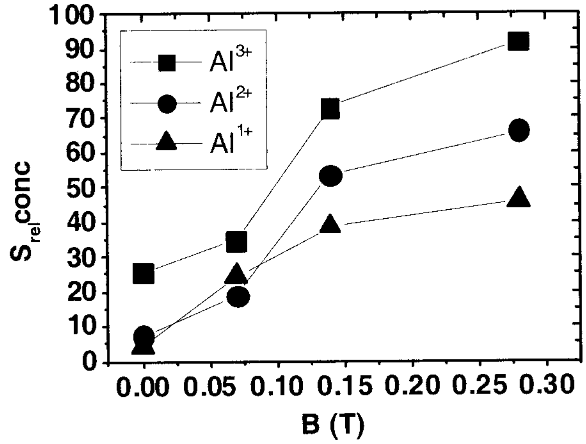

FIG. 1. Relative standard deviation of aluminum ion concentrations vs magnetic field, each data point is based on 100 plasma pulses.

cathode potential with respect to the grounded anode. It is known[6,7] that almost all of this voltage drops in the very thin cathode fall, thus the burning voltage is directly proportional to the power available for cathode spot operation. We again performed a statistical analysis of 100 pulses per parameter set. We found that an increase in magnetic field strength causes an increase in (a) the average burning voltage as well as (b) the fluctuations of the burning voltage, which are shown in Fig. 2. Since the arc current is kept constant this implies that (a) the power invested in the cathode surface is increasing with increasing B-field and (b) the fluctuation of the power is also increased with increasing B. These fluctuations are due to the well-known nonstationary character of cathodic arc spots,[6,7] i.e., the rapid formation and extinction of plasma production centers.

As the power invested in the cathode spot increases, the electron temperature increases,[8] which in turn affects the plasma composition. The correlation of electron temperature and plasma composition for an equilibrium plasma is given by the Saha equations.[9] The model of partial local Saha equilibrium[10] (PLSE) implies that the expanding plasma can be divided in distinct zones. In the first zone very close to the cathode spot, the plasma density is high enough to provide for sufficiently frequent collisions between electrons, atoms, and ions to result in ionization equilibrium, and thus the ratios between differently charged particles can be described by a set of Saha equations (Saha equilibrium). As the plasma expands, the rate of ionizing and recombining collisions decreases due to decreasing plasma density. The plasma reaches a temperature and density dependent transition zone, characterized by ionization equilibrium between ions of neighboring charge states that are separated by relatively small ionization energy, while the ionization and recombination rates of ions with high ionization energy are not equal due to insufficient frequency of inelastic collisions. The ion charge state ratios freeze successively in the expanding plasma, where the ionization transition with the largest ionization energy freezes first. After the plasma has gone through the PLSE transition zone, the charge state distribution (CSD) remains practically constant when the plasma is further expanding ("frozen distribution"). The PLSE model is verified for *vacuum* arc discharges,[9,10] and later we show that our findings for the metal species of arcs in reactive gases are qualitatively consistent with plasma expansion described by the PLSE freezing model.

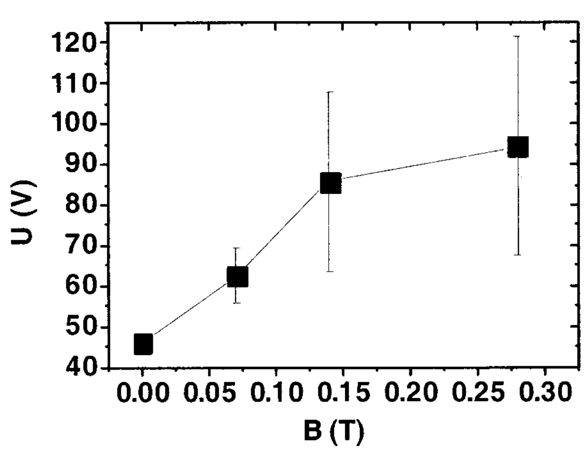

FIG. 2. Average burning voltage vs magnetic field, the bars represent standard deviation based on 100 plasma pulses per measuring point.

Fluctuations of the burning voltage represent fluctuations of the power invested in the plasma, and therefore the plasma parameters such as the electron temperature exhibit fluctuations as well. The ratios of ion concentrations are coupled to plasma density and electron temperature. Therefore, *fluctuations* of electron temperature must be associated with *fluctuations* of the freezing conditions in the transition zone and the resulting charge state distribution. A magnetic field increases the range of fluctuations (Fig. 1). The reason for the increase of fluctuations is not precisely known but it is reasonable to consider that the step distance between explosion centers is increased,[6,7] thereby ion assistance in spot formation is reduced. This would cause the impedance to go through larger amplitude variations. Impedance fluctuations are more likely to be amplified in magnetized plasmas than in nonmagnetized plasmas.[11] In addition to causing larger fluctuations, the magnetic field also causes electron impact ionization of the oxygen ambient[5] and residual gas[5,12] in the vacuum chamber.

The plasma composition is important since it is strongly correlated to the ion energy of the impinging ions during film growth. An expression for the kinetic energy of an ion is given by $E_i = E_0 + Qe\Delta U$, where E_0 is the initial kinetic energy of the ion gained at the cathode spot, Q is the charge state of the ion, and ΔU is the potential difference in the sheath in front of the substrate. Through ΔU, the ion energy can be controlled by selecting the substrate bias; however, the fluctuations discussed here about the plasma chemistry result in an extensive variation in ion energy. For example, if $\Delta U = 500$ V and the average charge state $\langle Q \rangle = 2$ with a variation of $\pm 10\%$, the average kinetic ion energy will accordingly have a variation of ± 100 eV. Therefore, the fluctuation of the CSD will affect the resulting microstructure of the film.

In summary, an increasing magnetic field causes an increase in the pulse-to-pulse fluctuations of aluminum ion concentrations in a reactive environment. This is consistent with the model of PLSE, suggesting that the fluctuations in plasma chemistry are caused by fluctuations of the plasma parameters in the cathode spot and transition zone to nonequilibrium. The plasma fluctuations in turn are the result of a fluctuating power density due to a fluctuating burning voltage. The resulting CSD may have a profound influence on the properties of the film since it is strongly connected to the ion energy, which is well-known to affect the microstructure evolution.

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