DIRECT VISUAL OBSERVATION OF POWDER DYNAMICS IN RF PLASMA-ASSISTED DEPOSITION

A.A. Howling, Ch. Hollenstein and P.J. Paris

DIRECT VISUAL OBSERVATION OF POWDER DYNAMICS IN RF PLASMA-ASSISTED DEPOSITION

A. A. Howling, Ch. Hollenstein and P.-J. Paris

Centre de Recherches en Physique des Plasmas, École Polytechnique Fédérale de Lausanne,

Av. des Bains 21, 1007 Lausanne, SWITZERLAND

ABSTRACT

Contamination due to particles generated and suspended in silane rf plasmas is investigated. Powder is rendered visible by illumination of the reactor volume. This simple diagnostic for global, spatio-temporal powder dynamics is used to study particle formation, trapping and powder reduction by power modulation.

Particulate contamination is of great concern in plasma-assisted deposition. The plasma itself is now recognised as a source of particles [1,2] and so in-situ powder diagnostics are needed to help resolve this problem. Laser scattering experiments measure transient particle passages across the laser path and spatial information is obtained by displacing the laser or the reactor. A global view of powder in etching plasmas (where particle number densities are much lower) was demonstrated using a rastered laser [3]. In this paper we simply illuminate the reactor volume, whereby global, spatio-temporal powder dynamics and particle trajectories are rendered visible; no moving mirrors, lasers or reactors are necessary. These preliminary experiments make no claim to be quantitative, but nevertheless give an immediate insight into powder dynamics.

The parallel plate reactor has two steel electrodes of diameter 135mm with 20mm separation, and the gas inlet is in the reactor wall. A halogen lamp served as the white light source and the powder structure was recorded on video.

The usual pure silane discharge image is the blue emission of the SiH* molecular band around 414nm. Fig. 1 shows a plasma illuminated from the left with white light. Filtering out the plasma emission around 414nm would leave a picture of powder alone. The video film shows a uniform stream of powder flowing radially from the layers past the electrode edge, forming a yellowish band around the reactor wall. The electrodes are protected from particle contact by the sheaths. Powder formed at the lower electrode edge by the fringing field between the earth screen and the rf electrode surface leaves the discharge volume directly. Attempting to reduce powder production by installing lateral screens to improve electric field homogeneity [4] could aggravate contamination by blocking the natural powder exhaust route.

On plasma extinction the powder is ejected laterally, but some settles on the lower electrode; hence the substrate is placed on the underside of the upper electrode. If the reactor was not discharge-conditioned, or the plasma power too high, the plasma emission flickers. On illumination, filaments of powder are visible near the electrode vertical axis; powder formed at the instant of ignition extinguishes the plasma which re-strikes elsewhere on the electrode surface. In some circumstances a plume of powder stretches from the electrodes back to the gas inlet in the reactor wall: illumination is useful for revealing unexpected powder sources.

Particles collected from the lower electrode, which were unoxidised, were ≈0.3μm in diameter. Thus visible light is Mie-scattered and the intensity is a convolution of the particle number density, size distribution, refractive index and wavelength. In-situ estimation of the particle number density and size is therefore difficult even with a laser source [2]. Another method, also requiring extensive analysis, measures the distribution of scattered light fluctuations [5]. Nevertheless, the scattered intensity of white light is a guide to the quantity of powder in the plasma, and we believe that here the convenience outweighs the simplification of analysis to be gained by using other souces. Digitisation of the video image (Fig. 2) can be used to give, for example, a vertical profile of the scattered intensity as shown in the right-hand side of Fig. 3. Powder layers are formed at very low powers for electrodes at 300K; at high powers, larger particles 'snow' down onto the lower electrode. For hot electrodes (500K) there is a power threshold below which no visible powder is formed and the matching condition is independent of power. If the power is increased up to the threshold, the matching begins to change. At this instant, no powder can be seen. Subsequently, the evolution of the powder profile becomes visible; Fig. 3 shows a representation in time. The reflected power measured before the matching box increases almost linearly with time until the layer reaches steady-state (Fig. 3). These observations relate to the accumulation of *visible* particles and do not necessarily indicate the spatial origin of nucleation. Their motion is probably determined by the negative charge achieved at floating potential. The initial phase of powder formation produces macromolecules or 'clusters' which, although invisibly small, introduce electron Coulomb scattering, attachment and recombination centres [6] and alter the electron energy distribution function [7]. These molecular-scale reactions modify the plasma impedance and matching condition. A change in reflected power is thus a *precursor to visible particle formation* which is useful because even 20Å particles could be deleterious to the electronic properties of a growing layer [4]. The reflected power could be incorporated into a feedback loop to keep the input power below the threshold for particle production, thus automatically maximising deposition rates without risk of contamination.

Powder has been filmed during the deposition of high-quality amorphous silicon with substrates heated to 500K despite the strong reduction of powder at elevated temperatures. An

explanation for this powder reduction could be that the radical sticking coefficients change sufficiently with substrate temperature to alter the radical composition in the discharge. Supporting evidence is the fact that the deposition rate is a strong function of substrate temperature [8]. If radicals responsible for powder formation have a higher sticking coefficient at higher substrate temperature, they would be removed from the discharge before agglomeration into clusters could begin. Another observation which suggests that powder production is linked to surface radical reactivity is the appearance, at high plasma powers, of a powder pattern on the lower steel electrode which is the reflection of the steel template holding the square glass substrate on the upper electrode. It would seem that, in the early stages of deposition, the different sticking coefficients for glass and steel cause different powder production rates, resulting in the pattern formed on the lower electrode.

To investigate powder trapping without continuous production, the silane gas flow was cut from a silane/hydrogen plasma; powder formation was thus arrested and the hydrogen plasma acted as a carrier discharge. Powder could be seen under illumination, showing that the negatively-charged particles are trapped in shallow, closed potential wells. The vertical retaining electric field is due to the sheath and the particles are perhaps attracted to this interface due to a polarisation of the charge cloud surrounding the particle [9]. The radial field is probably the ambipolar field equalising electron and positive ion diffusion. Powder streaming can thus be interpreted as a potential well overflowing due to mutual repulsion when the charged particle concentration becomes too large for the well to remain closed. Once a charged particle is trapped in a potential well its true residence time is larger than that calculated from gas flow rates. For low production rates, powder 'waves' can appear, slowly growing and then suddenly ejecting powder laterally from the discharge with simultaneous oscillations in the reflected power and electrode voltage: charged particles accumulate until they overflow as for the streaming case, but here the potential well is sufficiently perturbed by the charge loss that the ambipolar confinement collapses with a momentary loss of particles; the cycle then repeats every few minutes. These particle trapping phenomena suggest methods for powder suppression: for example, two biased ring electrodes at the height of the powder layers, slightly larger than the electrode radius, could perturb the time-averaged ambipolar field sufficiently to

cause negatively-charged clusters to fall onto their surface, thus preventing any build-up of larger particles by reducing the cluster residence time to a minimum. Electrical control of the residence time followed by extraction of powder of the required size would have interesting applications for ceramics, especially considering the high purity of powders produced in low pressure, cold plasmas [10]. Powder reduction by modulation of the rf power [11] is demonstrated in Fig. 4, where the plotted intensity is integrated over a vertical profile at the substrate position. At 1kHz, the only remaining visible powder is due to the rf electrode fringing fields which leaves the electrode region directly.

In summary, illumination of the discharge in plasma deposition reactors with visible light gives immediate insight into the problem of particulate contamination. Global, spatio-temporal motion of powder and particle trajectories can be recorded on video. Implementation is particularly simple. A change in rf matching gives a practical method for detecting the onset of cluster production. Illumination is especially suited to powder synthesis reactors, where powder streaming contours could be followed in order to optimise electrode design and powder collection.

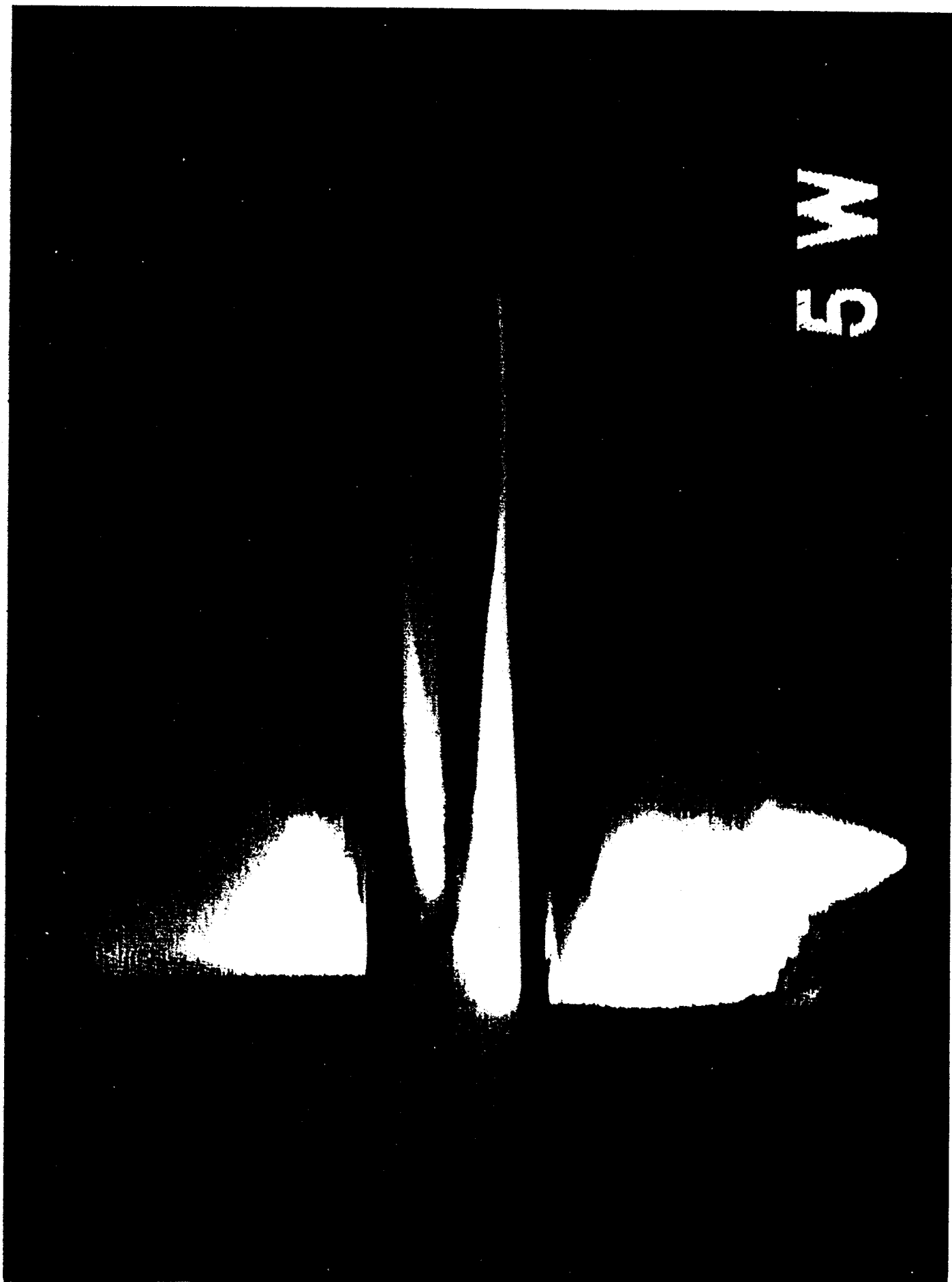## REFERENCES

1   G. S. Selwyn, J. Singh and R. S. Bennett, J. Vac. Sci. Technol. A7, 2758 (1989)

2   K. G. Spears, T. J. Robinson and R. M. Roth, IEEE Trans. Pl. Sci. PS-14, 179
     (1986)

3   G. S. Selwyn, J. E. Heidenreich and K. L. Haller, Appl. Phys. Lett. 57, 1876 (1990)

4   A. Gallagher, Int. J. Solar Energy, 5, 311 (1988)

5   K. G. Spears and T. J. Robinson, J. Phys. Chem. 92, 5302 (1988)

6   A. Garscadden, 'Nonequilibrium Processes in Partially Ionised Gases', Edited by M.
     Capitelli and J. N. Bardsley, Plenum Press, NY, 541 (1990)

7   M. J. McCaughey and M. J. Kushner, Appl. Phys. Lett. 55, 951 (1989)

8   F. Finger , U. Kroll, V. Viret, A. Shah, W. Beyer, X.-M. Tang, J. Weber, A. Howling
     and Ch. Hollenstein, submitted to J. Appl. Phys.

9   A. Bouchoule, A. Plain, L. Boufendi, J.Ph. Blondeau and C. Laure, submitted to
     J. Appl. Phys.

10   H. M. Anderson, R. Jairath and J. L. Mock, J. Appl. Phys. 67, 3999 (1990)

11   Y. Watanabe, M. Shiratan, Y. Kubo, I. Ogawa and S. Ogi, Appl. Phys. Lett. 53, 1263
     (1988)

FIGURE CAPTIONS

FIG. 1   Video image of an illuminated plasma with a 30sccm flow of silane at 0.3mbar, 5W, 60MHz, electrode temperature 300K. The viewing angle obscures the right-hand electrode edge. The stationary powder layers are indistinguishable without illumination.

FIG. 2   Digitisation of the video image showing the spatial distribution of the scattered light intensity. The heavy lines mark the electrode edges.

FIG. 3   Right-hand side: Vertical intensity profile at the substrate position of the light scattered from the powder layers. Left-hand side: Schematic of the formation of the powder layers and the variation of reflected power with time.

FIG. 4   Effect of power sine-wave modulation frequency on the intensity of light scattering (peak power = 20W).
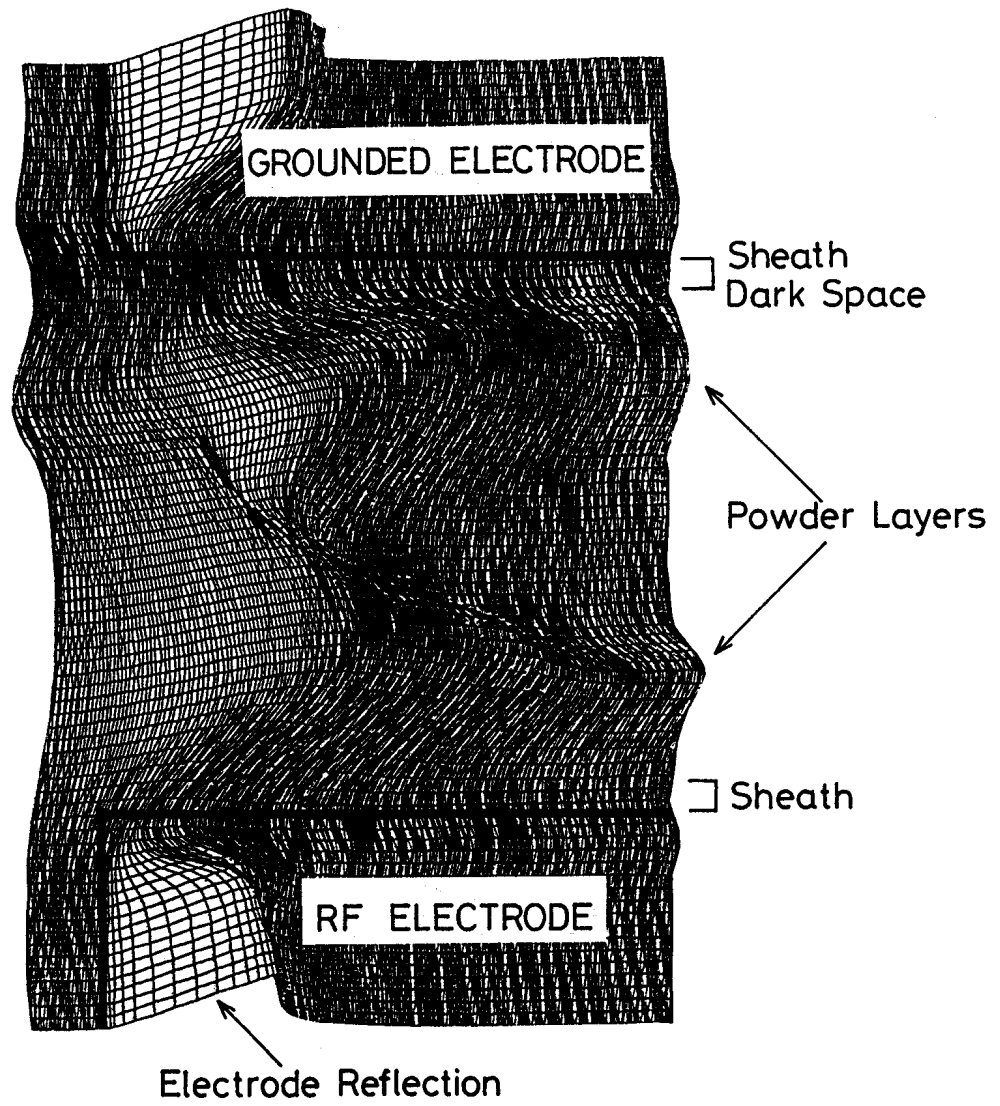
Fig. 1

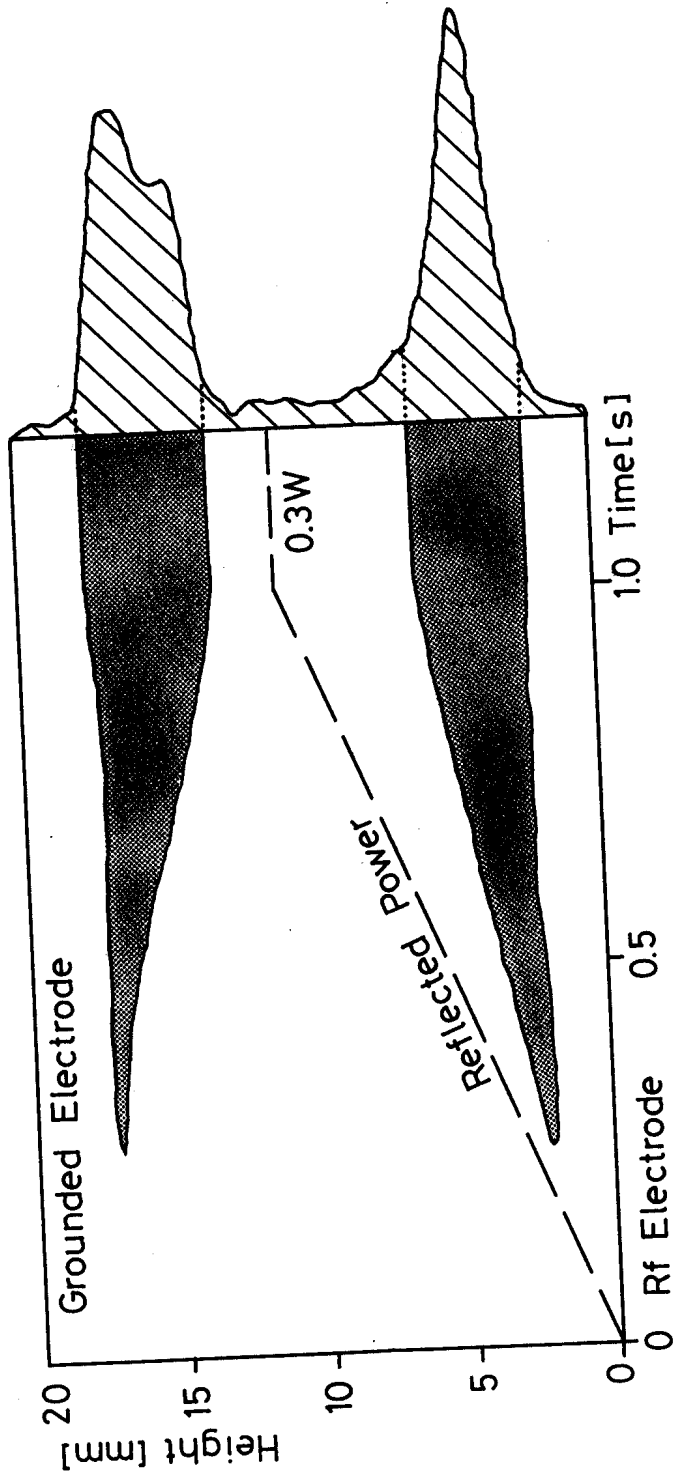GROUNDED ELECTRODE

⌐ Sheath
Dark Space

Powder Layers

⊐ Sheath

RF ELECTRODE

Electrode Reflection

Fig. 2

Fig. 3

Fig. 4