Physics of Collisional Plasmas

Introduction to high-frequency discharges

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Preface

During the 1960s, the public face of plasma physics was almost exclusively represented by plasma confinement, with the goal of developing a reactor to produce electricity by thermonuclear fusion. Such a reactor is still being developed, without any guarantee as to its successful achievement, but since then the applications of plasma physics have increased and diversified: one of the best known, besides lighting, is etching in the fabrication of microelectronic computer chips, for which plasma is indispensable. At present, the use of plasmas continues to expand and, from recent research publications, a seemingly limitless number of applications will eventually see the light of day. In this development, plasmas created by radiofrequency and microwave fields play a particularly important role.

The present text is basically concerned with plasma physics of interest for laboratory research and industrial applications, with emphasis on the understanding of the physical mechanisms involved, rather than on minute details and high-level theoretical analysis. At the introductory level to this discipline, it is very important to assimilate its characteristic physical phenomena, before addressing the ultimate formalism of kinetic theory, with its microscopic, statistical mechanics approach. In this textbook, the physical phenomena have been translated into more tractable equations, using the hydrodynamic model; this treats the plasma as a fluid, in which the macroscopic physical parameters are the statistical averages of the microscopic (individual) parameters. This textbook is intended for students in their early years at the graduate level, and for engineers who are interested in applications. Its level of difficulty lies somewhat below that of JL Delcroix and A Bers (from Université Paris XI, Orsay and Supélec, Gif-sur-Yvette, France, and MIT, Cambridge, MA, USA, respectively), which provides a series of complementary and interesting theoretical treatments.

This book is divided into four chapters.

Chapter 1 is the introductory part of the textbook. It begins with a description of the plasma, an ionised gas, as a collective and electrically neutral gaseous medium, followed, for illustrative purposes, by a few selected scientific and industrial applications. Then, the fundamental concepts of plasma physics are introduced, with progressively increasing detail: the chapter aims to present the basic parameters required to reach a starting knowledge of the plasma medium, such as the Debye length, the electron plasma frequency, the various types of collision between particles and their description through specific cross-sections. The concepts presented in this introduction will be developed as a first approach, i.e. as the first step in an iterative process, to be completed by the detailed and quantitative presentations in the remaining chapters.

Chapter 2 is a thorough examination of the trajectory of a single charged particle (assuming no interaction whatsoever with other particles), subject to an electric field $\boldsymbol{E}$, a magnetic field $\boldsymbol{B}$ or both. In the case of electric fields $\boldsymbol{E}$, special attention will be paid to those at RF and microwave frequencies, designated jointly as high-frequency (HF) fields, in preparation for the modelling of HF discharges developed in Chap. 4. The presence of a magnetic field $\boldsymbol{B}$ results in a cyclotron motion, encountered for example in electron cyclotron-resonance discharges (Chap. 4). The combination of $\boldsymbol{E}$ and $\boldsymbol{B}$ fields in different spatial configurations, and then the inclusion of the spatial inhomogeneity of the $\boldsymbol{B}$ field, reveals the so-called drift velocities, which have to be "tamed" for an efficient operation of Tokomaks, nowadays investigated as possible controlled-fusion reactors.

In contrast to Chap. 2, collisions between particles are taken into account in **Chapter 3**, to establish the hydrodynamic description of the plasma, considered as a fluid. Such a description is obtained from the macroscopic quantities calculated from the distribution function of the (microscopic) velocities of individual particles. The transport equations, i.e. the equations describing the space-time evolution of these quantities, are obtained from integration of the Boltzmann equation over the distribution function of velocities. The concepts of mobility (of charged particles) and diffusion of particles are then introduced, where free mobility and diffusion tensors are deduced from the (momentum transport) Langevin equation. Further, it is shown that, under sufficiently dense plasma conditions, the space-charge electric field makes electrons and ions diffuse together in the so-called ambipolar diffusion regime. Finally, toward the end of the chapter, a first example of a scaling law in plasmas is developed. Then, in the last section, the formation of sheaths located at the interface between the plasma and the walls is described, together with a straightforward and original derivation of the Bohm Criterion, which provides the velocity of the ions as they enter the sheath.

Chapter 4, the last chapter, is dedicated to the mechanisms involved in HF sustained discharges, which are developed based on an entirely new and original approach. The key element is θ_l, the average power lost by an electron through its collision with heavy particles, in this way supplying power to the plasma. It is shown that θ_a, the power taken on average per electron from the HF field, adjusts so that $\theta_l = \theta_a = \theta$, i.e. to compensate for the loss of charged particles. This implies, for instance, that the intensity of the

E field in the plasma is not set by the operator, but by this balance requirement. A further consequence is that, for given operating conditions and HF power density, whatever the means of supplying the HF field to achieve the discharge, the θ value should be the same in all cases. The parameter θ is also instrumental in demonstrating that, contrary to common belief, the ***E***-field intensity goes through a minimum at electron cyclotron resonance. The influence of varying the field frequency on the EEDF, and ultimately on plasma properties, is documented both theoretically and experimentally, in the case of low-pressure ($< 10\,$torr) plasmas. The case of high-pressure plasmas (including atmospheric pressure) is centred on the phenomena of discharge contraction and filamentation in rare gases with low thermal conductivity, emphasising the role of molecular ions in these monoatomic gas discharges. Interrupting the kinetic cycle leading to dissociative recombination (of molecular ions) by introducing traces of rare gases with an ionisation potential lower than that of the carrier gas leads to the disappearance of discharge contraction and filamentation.

In addition to the content of the main text, there are a large number of remarks and footnotes, for clarification, or to qualify certain points more precisely. Forty five problems, with detailed solutions, which are an indispensable complement to this book, are distributed at the end of the first three chapters. A set of Appendices provides clarifications of the subjects treated in the main text, together with a number of mathematical developments, and useful mathematical formulae. Finally, an alphabetic index of important terms is supplied, with a page reference to their first appearance in the text given in bold type.

Montréal and Grenoble, *Michel Moisan*
January 2012 *Jacques Pelletier*

1.2.5 Surface treatment

Plasma surface treatment consists of modifying the state of a surface by one of the following generic methods:

- *deposition* of a thin film of a given material (metal; semi-conductor; dielectric; polymer) on the surface;
- *chemical reaction* with the surface itself (oxidation; nitriding) or *physico-chemical* transformation of the surface (modification of adherence, surface energy);
- *erosion* of the surface, either by a chemical reaction, which involves the formation of a volatile molecule, from one or more atoms from the surface and atoms or radicals provided by the plasma, or a physical action, *sputtering* by ion bombardment, such that ions eject atoms from the surface by a mechanical process, or by a chemical reaction assisted (induced) by ion bombardment, which combines chemical and physical actions.

Thus, a plasma produced from the gas CF_4 creates, in the volume, atoms (such as F), radicals (such as CF_x) as well as ions (such as CF_y^+) and more complex species necessary for interactions with the surface that, under suitable operating conditions, can equally well lead to etching of materials (Si, W, SiO_2) as is illustrated in Fig. 1.4, or deposition of teflon-like thin films by plasma-induced polymerisation. In the fabrication of micro-electronic chips, due to the requirements of smaller and smaller miniaturisation, the use of plasma continues to expand its range of applications: *surface cleaning, etching* (production of "patterns" in the substrate by surface erosion), deposition, *ion implantation* (doping by introducing ions deep in the material), *lithography* (impression and "photographic" development of resins allowing the transfer of patterns to define electric circuits), oxidation and thermal treatments.

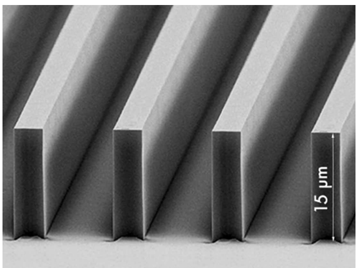

Fig. 1.4 Example of anisotropic etching of SiO_2 (courtesy of CORIAL, France).

Of the multitude of elementary steps required for the fabrication of integrated circuits, the operations uniquely realised by plasmas represented, at the beginning of the 2000s, close to 50% of the total number of these steps. The introduction of plasma equipment for micro-electronics, and more gener-

Electron-neutral collisions leading to the ionisation of an atom (molecule)

In laboratory plasmas, ionisation of a gas usually occurs as a result of electron impact. The probability of impact ionisation by atom-atom collisions is, in fact, small in plasmas with less than a few atmospheres pressure. If we take an argon atom, its step-wise ionisation through the metastable states as a relay, which is the lowest energy pathway to achieve ionisation, requires a minimum energy of 11.5 eV (Appendix VI). Thus, assuming a Maxwell-Boltzmann particle energy distribution function, this would require that the most energetic atoms in the plasma reach a temperature of more than 100,000 K, which is not realistic in laboratory plasmas.

The ionisation cross-section for electron-neutral collisions generally exhibits the following characteristics (Fig. 1.15):

- a very precise energy threshold $\mathcal{E}_i$, below which the cross-section is zero. For atoms, the *ionisation threshold* corresponds to the *ionisation potential*. For molecules, a number of ionisation thresholds co-exist (dissociative and non dissociative ionisation).
- immediately above the energy threshold $\mathcal{E}_i$, an (almost) linear growth of the cross-section as a function of the energy U_{eV} of the electron[40],
- then the cross-section passes through a maximum for an energy $\mathcal{E}_m$, followed by a slow diminution.

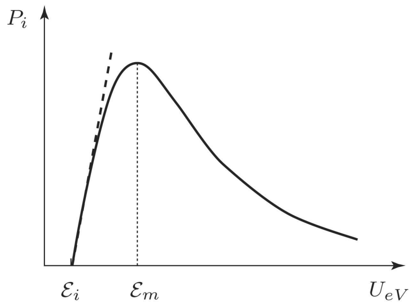

Fig. 1.15 General form of the ionisation cross-section of an atom by electron collisions.

The dashed line in Fig. 1.15, with slope a_i, drawn from the energy threshold $\mathcal{E}_i$ of the cross-section, represented by the expression:

$$P_i = a_i(U_{eV} - \mathcal{E}_i) \text{ for } U_{eV} \geq \mathcal{E}_i \ , \tag{1.142}$$

is a good approximation for the initial portion ($\mathcal{E}_i < U_{eV} < \mathcal{E}_m$) of the ionisation cross-section.

[40] Since the target-particles are not compelled to be at rest, it is more correct to speak of their relative energy (velocity) at the moment of collision. In the case of electron-neutral interactions, this distinction is generally negligible above a fraction of an electron volt in the case where the gas is not very warm (see also remark 2 at the end of this section).

Characteristic frequency ν_{Dk} of the different modes: relative values compared to that of the fundamental mode

We wish to evaluate the relative contributions to electron density of the various diffusion modes appearing in the sum (3.225); to do this, we extract from Eqs. (3.226) and (3.227) for $k = 1$, $\nu_{D1} = D\pi^2/L^2$; for $k = 2$, $\nu_{D2} = 9\,D\pi^2/L^2$; for $k = 3$, $\nu_{D3} = 25\,D\pi^2/L^2$, such that $\nu_{D2}/\nu_{D1} = 9$, $\nu_{D3}/\nu_{D1} = 25$, etc. Then, it is clear that the fundamental mode decays the least rapidly in (3.225): no matter what the initial spatial density distribution was (obtained, for example, as a result of a laser pulse focussed at a point inside the discharge vessel), after a certain time, it will take the shape of the fundamental mode, with the other terms in (3.225) making no significant contribution, as is illustrated in Fig. 3.2.

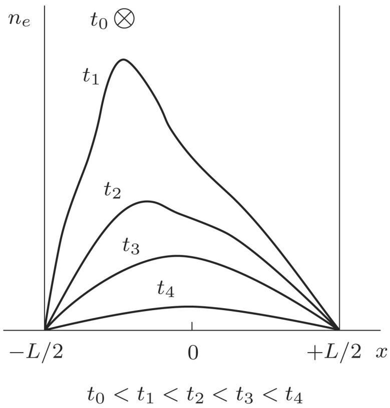

Fig. 3.2 Approximate time evolution of the electron density, in the diffusion regime, of a plasma created locally ($\otimes$) at time t_0.

Diffusion length

For the fundamental mode ($k = 1$), we have $\dfrac{L}{2\Lambda} = \dfrac{\pi}{2}$, from which:

$$\Lambda = L/\pi \, , \tag{3.228}$$

where Λ appears as a characteristic diffusion length in the planar configuration.

Cylindrical configuration

We will consider a cylindrical vessel, closed at each end by planar surfaces (Fig. 3.3).

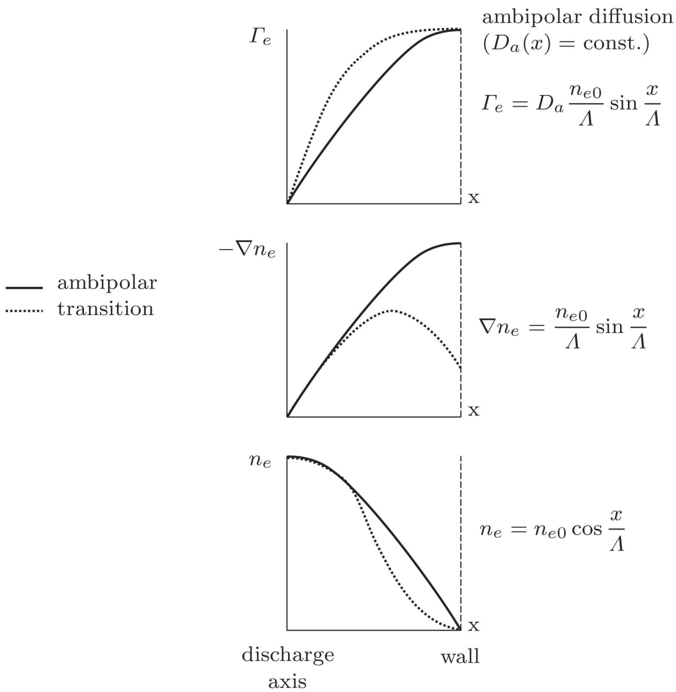

Fig. 3.6 Numerically calculated values of Γ_e and n_e as functions of spatial position x in planar geometry, compared to those obtained when assuming a constant ambipolar diffusion coefficient: the calculated diffusion coefficient changes from truly ambipolar on the axis ($x = 0$) to that of the transition regime as density decreases towards the wall (after Brown).

Density – diffusion length criterion

We can generally say that the field intensity $\boldsymbol{E}_D$ required for ambipolar diffusion is reached when $\Lambda^2\sigma_0$ is sufficiently large (see (3.274)), or more specifically when:

$$n_{e0}\Lambda^2 > 10^7 \,\text{cm}^{-1} \,, \tag{3.276}$$

where n_{e0}, the electron density on the axis, is expressed in cm^{-3} and Λ in cm. For values of $n_{e0}\Lambda^2$ less than $10^5 \,\text{cm}^{-1}$, the plasma is in the free diffusion regime, provided that the conditions are actually those of a diffusion regime (collision mean free path $< R$) and not that of free fall (the definition of free fall will be discussed later, in Sect. 3.12).

Remarks:

1. The average electron density (over the cross-section) of a classical fluorescent lamp tube, maintained by an alternating current, varies between

3.17. In general, the characteristic diffusion length Λ of a plasma is related to plasma dimensions by coefficients that depend on the plasma geometry, (see Sect. 3.9.1), but also on the boundary conditions chosen. Therefore, we would like to calculate the characteristic diffusion length in a plasma for different geometrical configurations (planar and cylindrical) in the case where the assumption $n(r = R) = 0$ is no longer valid, i.e. when the losses to the walls of the experiment are through a flux across an ion sheath. Assume that the plasma is in the ambipolar diffusion regime and that the neutrality condition ($n_i = n_e$) is applicable up to the edge of the collisionless ion sheath, whose thickness can be neglected when compared to the plasma dimensions.

a) Calculate the characteristic diffusion length $\Lambda = L/\mathsf{a}$ (where a is a dimensionless coefficient) of a planar configuration that is infinite along y and z, and with a width along x of $L = 2\,\text{cm}$, for the two following cases:

1. Argon plasma, $p = 0.5\,\text{torr}$, $T_{eV} = 1.7\,\text{eV}$, $T_g = 300\,\text{K}$, mobility of Ar^+ ions in Ar: $\mu_{i0} = 1.52\,\text{cm}^2\,\text{V}^{-1}\,\text{s}^{-1}$ at 760 torr and 273 K.
2. Helium plasma, $p = 0.5\,\text{torr}$, $T_{eV} = 5.8\,\text{eV}$, $T_g = 700\,\text{K}$, mobility of He^+ ions in He: $\mu_{i0} = 10.4\,\text{cm}^2\,\text{V}^{-1}\,\text{s}^{-1}$ at 760 torr and 273 K.

To calculate a, use the graph $\mathsf{a} \tan \mathsf{a} = f(\mathsf{a})$.

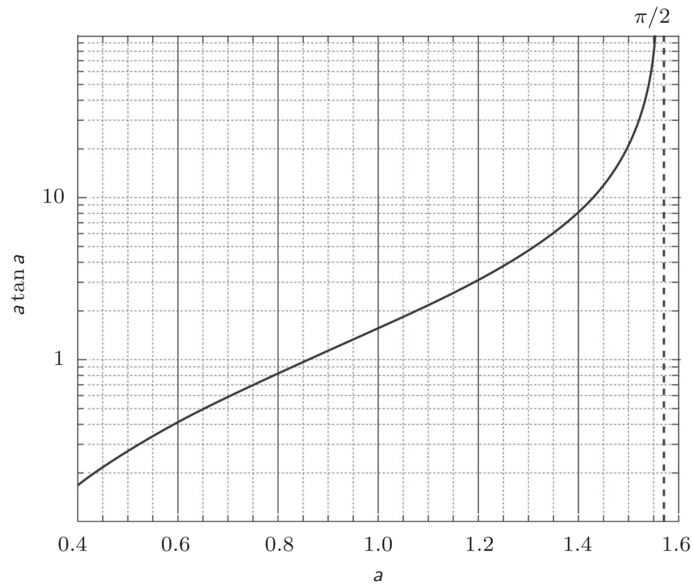

Fig. 3.14 Variation of the function $f(\mathsf{a}) = \mathsf{a} \tan \mathsf{a}$ with a.

b) Calculate the characteristic diffusion length $\Lambda = L/\mathsf{b}$ (where b is a dimensionless coefficient) of an infinite cylindrical plasma column of radius $R = 1\,\text{cm}$ for the two sets of plasma conditions defined above.

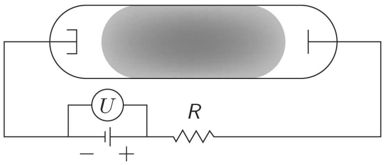

Fig. 4.1 Schematic of an electric discharge maintained at constant current, usually referred to as a direct current (DC) discharge. The resistance R (ballast) ensures the stability of the discharge.

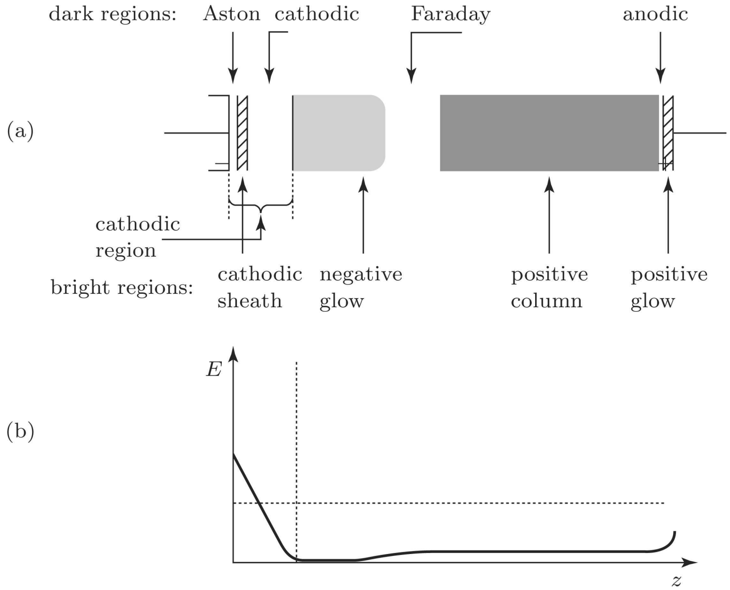

Fig. 4.2 **a** Representation of the different dark and luminous zones in a direct current discharge, together with **b** the qualitative variation of the electric field intensity E in the stationary state, along the length of the discharge. The horizontal dashed line indicates the electric field intensity E before ignition.

To characterise the transfer of power from the field $\boldsymbol{E}$ to the positive column plasma by means of the electrons, we will establish the balance between the power taken (on average) by an electron from the electric field, referred to as the absorbed power θ_a (Sect. 2.2.1), and the power that the electron (on average) transfers to the heavy particles as a result of collisions, referred to as the power lost θ_l.

The average *power* θ_l *lost per electron*, and transferred to the plasma following the various types of collision of electrons with heavy particles [144] can be written [26]:

[144] Equation (4.1) can be obtained from the homogeneous Boltzmann equation by considering the isotropic part $F_0(U)$ of the EEDF. This equation is then multiplied by U and integrated over all values of U. The integral of $F(U)$ is described in Appendix XVII (see note at the bottom of the page).

Effect of the transition from a non stationary EEDF to a stationary EEDF on polymer deposition

Figure 4.16 shows the variation of the deposition rate of thin films on polymers, normalised to P_t, the total HF power absorbed[170], as a function of the frequency of the applied HF field. These coatings have been obtained from C_4H_8 (isobutylene) or C_4F_8 (perfluorocyclobutane) using argon as the carrier gas, the supply of the monomer being (under standard temperature and pressure conditions) 3 standard cubic centimeter/sec (sccm) and that of argon 10 sccm, with a total pressure of 0.2 torr [8]. The transition to the upper plateau for $f \geq 100$ MHz corresponds to the transition, for a stationary EEDF, from the DC case to that of a Maxwellian [24]. However, taking into account the presence of molecules in the discharge (which cause the stationary EEDF to be reached at a higher frequency than for atomic gases), one might then think that this involves the transition from a non-stationary EEDF to a stationary EEDF (DC case). Since we do not have such calculations on the EEDF at our disposal, it appears difficult to answer this question.

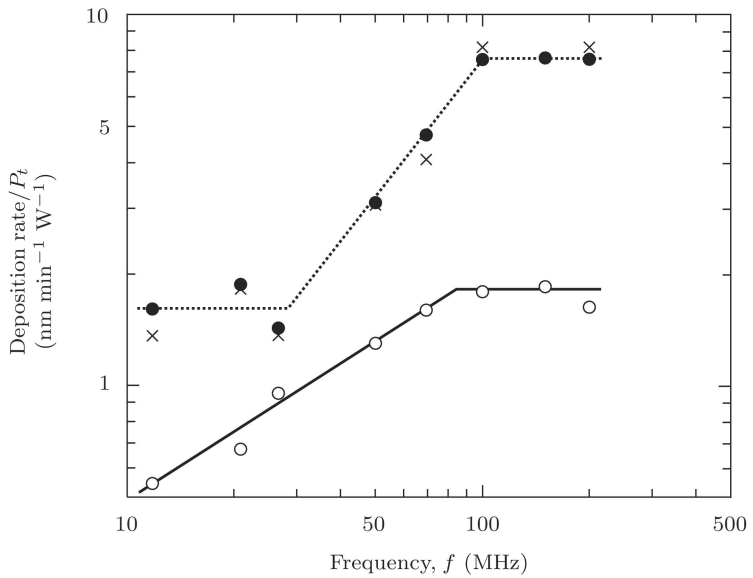

Fig. 4.16 Growth rate of polymer films, normalised to the total absorbed power P_t, as a function of the frequency of the applied HF field, $R = 30.5$ mm (○ C_4F_8, ● and × C_4H_8, for two values of P_t) [8].

[170] Since we are unable to maintain a constant power density in the present case, we normalise the deposition rate to P_t, the total power absorbed in the discharge.

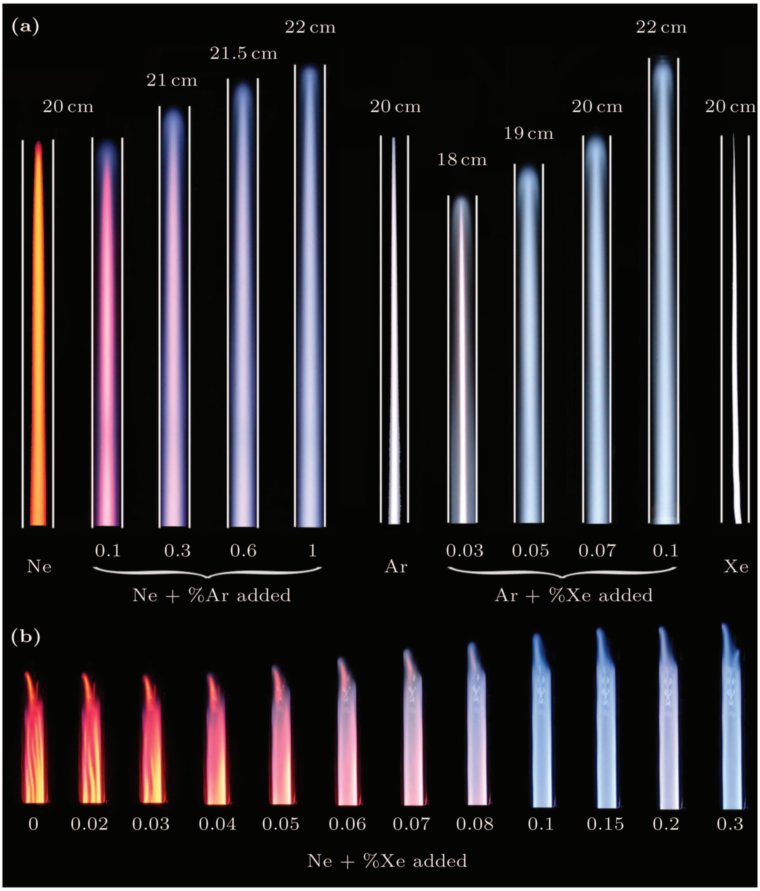

Fig. 4.21 Photographs of the upper section (with respect to the plane of the microwave field applicator, which is perpendicular to the tube axis) of a vertically oriented surface wave discharge in different noble gases at atmospheric pressure under a 0.5 slm gas flow: **a** for a tube of 12 mm inner diameter and a field frequency of 915 MHz, showing contraction in pure Ne and pure Ar. As traces of Ar are added to Ne and traces of Xe to Ar, there is a progressive expansion of the discharge; **b** for a tube of 20 mm inner diameter and a field frequency of 2450 MHz, showing filamentation. Progressively adding traces of Kr to Ne reduces filamentation, which ultimately vanishes [7].

case of a cylindrical discharge, the plasma, determined by its luminous section, occupies the total radial cross-section of the tube. This is related to the fact that the loss of charged particles (electrons and ions) takes place through diffusion to the walls of the tube where they recombine. Under these conditions, the radial distribution of electrons is determined only by the pressure and the radius of the discharge tube (Sect. 3.13). In contrast to the diffuse case, the electrons in a contracted discharge are confined to