Analysis of Instabilities in Non-Equilibrium Plasmas *

LIN Lie(林烈)[1][**], WU Bin(吴彬)[1], ZHANG Peng(张澎)[2], WANG Yong-Qing(王永庆)[2]

[1]*Institute of Mechanics, Chinese Academy of Sciences, Beijing 100080*
[2]*Shenyang Aircraft Research Institute, Shenyang 110035*

Plasma instabilities with charged particle production processes in non-equilibrium plasma are analysed. A criterion on plasma instabilities is deduced by first-order perturbation theory. The relationship between plasma instabilities and certain factors (degree of non-equilibrium in plasma, the electron attachment rate coefficient and electron temperature) are described.

It is well known that there have been many studies on instabilities of high-temperature plasmas. Some studies on the instabilities of low-temperature plasmas with attaching gases have been carried out, and some phenomena of instability in low temperature plasmas have been observed.[1−8] However, further research on instability in low-temperature plasmas, especially in non-equilibrium plasmas, is needed. In general, there are two types of instabilities in low-temperature plasmas: charged-particle production instability and thermal instability. The instability behaviour is usually manifested in the form of striations and/or constriction of plasmas. In this Letter, we study the first kind of instability and analyse the relationship between instability and certain factors in a plasma (degree of non-equilibrium of the plasma, rate coefficients of reaction in the plasma) with the first-order perturbation theory. Also, we deduce a criterion of instability in non-equilibrium plasmas.

We assume that the plasma medium is a flowing gas in the discharge chamber and it is an attaching gas. Then the conservation equations for electron and negative ions read

$$\frac{Dn_e}{Dt} + n_e \nabla \cdot U + \nabla \cdot (n_e U_e)$$
$$= n_e n k_i + n_n n k_d + n_n n_e k_d^e - n_e n_p k_r^e - n_e n k_a, \tag{1}$$

$$\frac{Dn_n}{Dt} + n_n \nabla \cdot U + \nabla \cdot (n_n U_n)$$
$$= n_e n k_a - n_n n_p k_r^i - n_n n k_d - n_n n_e k_d^e, \tag{2}$$

where $D/Dt = \partial/\partial t + U \cdot \nabla$, U is the velocity of gas flow, n_e is the electron density, n_n is the density of the negative ions, n is the density of the neutral particles, n_p is the density of positive ions, U_e and U_n are the diffusion velocities of the electron and the negative ion respectively; $k_i, k_d, k_a, k_d^e, k_r^e,$ and k_r^i are the electron production rate coefficient, detachment rate coefficient, attachment rate coefficient, detachment

rate coefficient for electron collision, the recombination rate coefficient for the electron and positive ions and the recombination rate coefficient for positive-ion and negative-ion respectively. For the flowing gas discharge plasma, the characteristic time of the gas flow is much longer than the time scale of the charged particle production and the space charges relaxation. Therefore, Eqs. (1) and (2) can be reduced to

$$\frac{\partial n_e}{\partial t} = n_e n k_i + n_n n k_d + n_n n_e k_d^e - n_e n_p k_r^e - n_e n k_a, \tag{3}$$

$$\frac{\partial n_n}{\partial t} = n_e n k_a - n_n n_p k_r^i - n_n n k_d - n_n n_e k_d^e. \tag{4}$$

If there are perturbations in the plasma, each of the densities n_e, n_n, and n_p and the charged-particle temperatures T_e and T_g can be considered to be composed of a steady-state value and a small spatially and temporally varying quantity, i.e.

$$n_e = \bar{n}_e + \tilde{n}_e, \quad n_n = \bar{n}_n + \tilde{n}_n, \quad n_p = \bar{n}_p + \tilde{n}_p,$$
$$T_e = \bar{T}_e + \tilde{T}_e, \quad T_g = \bar{T}_g + \tilde{T}_g, \tag{5}$$

where T_e and T_g are the electron temperature and the heavy particle temperature respectively. The first order perturbations in the rate coefficients have been expressed in terms of the electron temperature perturbations and heavy particles temperature perturbations, i.e.

$$k_i = \bar{k}_i + \frac{\partial k_i}{\partial T_e}\tilde{T}_e, \quad k_a = \bar{k}_a + \frac{\partial k_a}{\partial T_e}\tilde{T}_e,$$
$$k_r^e = \bar{k}_r^e + \frac{\partial k_r^e}{\partial T_e}\tilde{T}_e, \quad k_d^e = \bar{k}_d^e + \frac{\partial k_d^e}{\partial T_e}\tilde{T}_e,$$
$$k_d = \bar{k}_d + \frac{\partial k_d}{\partial T_g}\tilde{T}_g, \quad k_r^i = \bar{k}_r^i + \frac{\partial k_r^i}{\partial T_g}\tilde{T}_g. \tag{6}$$

The first-order equations for the perturbations in the electron and negative-ion densities are

$$\frac{\partial \tilde{n}_e}{\partial t} = \tilde{n}_e(n\bar{k}_i - \bar{n}_p\bar{k}_r^e - \bar{n}_e\bar{k}_r^e - n\bar{k}_a + \bar{n}_n\bar{k}_d^e)$$

$$+ \tilde{n}_n(nk_d - \bar{n}_e k_r^e + \bar{n}_e \bar{k}_d^e)$$

$$+ \tilde{T}_e \bar{n}_e \left(n \frac{\partial k_i}{\partial T_e} - \bar{n}_p \frac{\partial k_r^e}{\partial T_e} \right.$$

$$\left. - n \frac{\partial k_a}{\partial T_e} + \bar{n}_n \frac{\partial k_d^e}{\partial T_e} \right) + \tilde{T}_g \bar{n}_n n \frac{\partial k_d}{\partial T_g}, \qquad (7)$$

$$\frac{\partial \tilde{n}_n}{\partial t} = \tilde{n}_e(n\bar{k}_a - \bar{n}_n k_r^i - \bar{n}_n \bar{k}_d^e)$$

$$- \tilde{n}_n(\bar{n}_p \bar{k}_r^i + n\bar{k}_d + \bar{n}_n k_r^i + \bar{n}_e \bar{k}_d^e)$$

$$+ \tilde{T}_e \left(\bar{n}_e n \frac{\partial k_a}{\partial T_e} - \bar{n}_e \bar{n}_n \frac{\partial k_d^e}{\partial T_e} \right)$$

$$- \tilde{T}_g \left(\bar{n}_p \bar{n}_n \frac{\partial k_r^i}{\partial T_g} + \bar{n}_n n \frac{\partial k_d}{\partial T_g} \right). \qquad (8)$$

The perturbations in the plasma properties are represented in the usual way by a superposition of their Fourier components:

$$\tilde{n}_{ek} = n_{ek} e^{i\omega t}, \quad \tilde{n}_{nk} = n_{nk} e^{i\omega t},$$
$$\tilde{T}_{ek} = T_{ek} e^{i\omega t}, \quad \tilde{T}_{gk} = T_{gk} e^{i\omega t}. \qquad (9)$$

After some minor algebraic manipulations involving the steady state form of Eqs. (3) and (4), we can obtain

$$\frac{n_{ek}}{\bar{n}_e}(i\omega) = - \frac{n_{ek}}{\bar{n}_e} \left(\bar{n}_e \bar{k}_r^e + \frac{\bar{n}_n}{\bar{n}_e} n\bar{k}_d \right)$$

$$+ \frac{n_{en}}{\bar{n}_n} \left(\frac{\bar{n}_n}{\bar{n}_e} n\bar{k}_d - \bar{n}_n \bar{k}_r^e + \bar{n}_n \bar{k}_d^e \right)$$

$$+ \frac{T_{ek}}{\bar{T}_e} (nk_i \hat{k}_i - \bar{n}_p k_r^e \hat{k}_r^e - nk_a \hat{k}_a + \bar{n}_n k_d^e \hat{k}_d^e)$$

$$+ \frac{T_{gk}}{\bar{T}_g} \frac{\bar{n}_n}{\bar{n}_e} nk_d \hat{k}_d, \qquad (10)$$

$$\frac{n_{nk}}{\bar{n}_n}(i\omega) = \frac{n_{ek}}{\bar{n}_e} \left(\frac{\bar{n}_e}{\bar{n}_n} n\bar{k}_a - \bar{n}_e \bar{k}_r^i - \bar{n}_e \bar{k}_d^e \right)$$

$$- \frac{n_{nk}}{\bar{n}_n} \left(\frac{\bar{n}_e}{\bar{n}_n} n\bar{k}_a + \bar{n}_n \bar{k}_r^i \right)$$

$$+ \frac{T_{ek}}{\bar{T}_e} \left(\frac{\bar{n}_e}{\bar{n}_n} nk_a \hat{k}_a - \bar{n}_e k_d^e \hat{k}_d^e \right)$$

$$- \frac{T_{gk}}{\bar{T}_g} (\bar{n}_p k_r^i \hat{k}_r^i + nk_d \hat{k}_d), \qquad (11)$$

where

$$\hat{k}_i = \frac{\bar{T}_e}{k_i} \frac{\partial k_i}{\partial T_e} = \frac{\partial \ln k_i}{\partial \ln T_e}, \quad \hat{k}_d = \frac{\bar{T}_g}{k_d} \frac{\partial k_d}{\partial T_g} = \frac{\partial \ln k_d}{\partial \ln T_g},$$

$$\hat{k}_r^e = \frac{\bar{T}_e}{k_r^e} \frac{\partial k_r^e}{\partial T_e} = \frac{\partial \ln k_r^e}{\partial \ln T_e}, \quad \hat{k}_d^e = \frac{\bar{T}_e}{k_d^e} \frac{\partial k_d^e}{\partial T_e} = \frac{\partial \ln k_d^e}{\partial \ln T_e},$$

$$\hat{k}_a = \frac{\bar{T}_e}{k_a} \frac{\partial k_a}{\partial T_e} = \frac{\partial \ln k_a}{\partial \ln T_e}. \qquad (12)$$

The relationship between $\frac{T_{ek}}{\bar{T}_e}$ and $\frac{n_{ek}}{\bar{n}_e}$ is[5]

$$\frac{T_{ek}}{\bar{T}_e} = \left(\frac{-2\cos^2\phi}{1 + \hat{\nu}_u - \hat{\nu}_m \cos 2\phi} \right) \frac{n_{ek}}{\bar{n}_e} = \left(\frac{-2\cos^2\phi}{\hat{\nu}_u'} \right) \frac{n_{ek}}{\bar{n}_e}, \qquad (13)$$

where $\hat{\nu}_u$ and $\hat{\nu}_m$ are, respectively, the total electron energy exchange collision frequency and the momentum transfer collision frequency with the caret notation referring to logarithmic derivatives with respect to electron temperature, $\hat{\nu}_u = 1 + \nu_u - \hat{\nu}_m \cos(2\phi)$; ϕ is the angle between the direction of the steady state electric field and the wave propagation vector $\boldsymbol{k}$. We introduce the following relation:

$$\frac{T_{gk}}{\bar{T}_g} = \left(\frac{T_{gk}/\bar{T}_g}{T_{ek}/\bar{T}_e} \right) \frac{T_{ek}}{\bar{T}_e} = \eta \frac{T_{ek}}{\bar{T}_e}, \qquad (14)$$

where

$$\eta = \left(\frac{T_{gk}}{\bar{T}_g} \right) \Big/ \left(\frac{T_{ek}}{\bar{T}_e} \right). \qquad (15)$$

Thus we can obtain

$$(i\omega)^2 + \left[\left(\bar{n}_e \overline{k_r^e} + \frac{\bar{n}_n n}{n_e} \overline{k_d} \right) + \left(\bar{n}_n \overline{k_r^i} + \frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \right) \right.$$

$$+ \left(\frac{2\cos^2\phi}{\hat{\nu}_n'} \right) \left(n\bar{k}_i \hat{k}_i - \bar{n}_p \bar{k}_r^e \hat{k}_r^e - n\bar{k}_a \hat{k}_a \right.$$

$$\left. + \bar{n}_n k_d^2 \hat{k}_d^e + \eta \frac{\overline{n}_e n}{n_e} \bar{k}_d \hat{k}_d \right) \Big] (i\omega)$$

$$+ \left(\bar{n}_n \bar{k}_r^i + \frac{\bar{n}_e n}{n_n} \bar{k}_a \right) \left(\bar{n}_e \bar{k}_r^2 + \frac{\bar{n}_n n}{\bar{n}_e} \bar{k}_d \right)$$

$$+ \left(\bar{n}_n \bar{k}_r^i + \frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \right) \left(\frac{2\cos^2\phi}{\hat{\nu}_u'} \right)$$

$$\cdot \left(n\bar{k}_i \hat{k}_i - \bar{n}_p \bar{k}_r^2 \hat{k}_r^e - n\bar{k}_a \hat{k}_a + \bar{n}_n k_d^e \hat{k}_d^e \right.$$

$$\left. + \eta \frac{\bar{n}_n n}{n_e} \bar{k}_d \hat{k}_d \right) - \left(\frac{\bar{n}_n n}{n_e} \bar{k}_d - \bar{n}_n \bar{k}_r^e + n\bar{k}_d^2 \right)$$

$$\cdot \left\{ \left(\frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a - \bar{n}_e \bar{k}_r^i - \bar{n}_e \bar{k}_d^2 \right) + \left(\frac{2\cos^2\phi}{\hat{\nu}_u'} \right) \right.$$

$$\left. \cdot \left[\frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \hat{k}_a - \bar{n}_e k_d^e \hat{k}_d^e - \eta(\bar{n}_p \bar{k}_r^i \hat{k}_r^i + n\bar{k}_d \hat{k}_d) \right] \right\}$$

$$= 0. \qquad (16)$$

The two roots of Eq. (16) can be expressed in the form

$$i\omega = -\frac{b}{2} \pm \frac{1}{2}\sqrt{b^2 - 4c}, \qquad (17)$$

where

$$b = \left[\left(\bar{n}_e \bar{k}_r^e + \frac{\bar{n}_n n}{\bar{n}_e} \bar{k}_d \right) + \left(\bar{n}_n \bar{k}_r^i + \frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \right) \right.$$

$$+ \left(\frac{2\cos^2\phi}{\hat{\nu}_u'} \right) (n\bar{k}_i \hat{k}_i - \bar{n}_p \bar{k}_r^e \hat{k}_r^e - n\bar{k}_a \hat{k}_a$$

$$\left. + \bar{n}_n k_d^e + \eta \frac{\bar{n}_n n}{n_e} \bar{k}_d \hat{k}_d) \right], \qquad (18)$$

$$c = \left(n_n \bar{k}_r^i + \frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \right) \left(\bar{n}_e \bar{k}_r^e + \frac{\bar{n}_n n}{\bar{n}_e} \bar{k}_d \right)$$

$$+ \left(\bar{n}_n \bar{k}_r^i + \frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \right) \left(\frac{2\cos^2\phi}{\hat{\nu}_u'} \right)$$

$$\cdot \left(n\bar{k}_i \hat{k}_i - \bar{n}_p \bar{k}_r^e \hat{k}_r^2 - n\bar{k}_a \hat{k}_a + \bar{n}_n k_d^e \hat{k}_d^e \right.$$

$$\left. + \eta \frac{\bar{n}_n n}{\bar{n}_e} \bar{k}_d \hat{k}_d \right) - \left(\frac{\bar{n}_n n}{\bar{n}_e} \bar{k}_d - \bar{n}_n \bar{k}_r^e + n\bar{k}_d^e \right)$$

$$\cdot \left\{ \left(\frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a - \bar{n}_e \bar{k}_r^i - \bar{n}_e \bar{k}_d^e \right) \right.$$

$$+ \left(\frac{2 \cos^2 \phi}{\hat{\nu}_u'} \right) \left[\frac{\bar{n}_e n}{\bar{n}_n} \bar{k}_a \hat{k}_a - n_e k_d^e \hat{k}_d^e \right.$$

$$\left. \left. - \eta \left(\bar{n}_p \bar{k}_r^i \hat{k}_r^i + n \bar{k}_d \hat{k}_d \right) \right] \right\}. \tag{19}$$

The condition of production instability for the non-equilibrium plasma is

$$\omega > 0. \tag{20}$$

Careful consideration of the various physical processes contributing to the terms in the coefficients b and c shows that when $b < 0$, the negative ion mode and the ionization mode are unstable. Therefore, the criterion for the charged particle production instabilities can be expressed by

$$n k_i \hat{k}_i - \bar{n}_p k_r^e \hat{k}_r^e - n k_a \hat{k}_a + \bar{n}_n k_d^e \hat{k}_d^e + \eta \frac{\bar{n}_n n}{\bar{n}_e} k_d \hat{k}_d < 0. \tag{21}$$

In general, the density of the neutral particles is much higher than the density of the negative and positive particles in the low-temperature plasma, so criterion (21) can be simplified into

$$\frac{k_i \hat{k}_i}{k_a \hat{k}_a} + \eta \frac{\bar{n}_n}{\bar{n}_e} \frac{k_d \hat{k}_d}{k_a \hat{k}_a} < 1. \tag{22}$$

Criterion (22) indicates that if there is greater attachment rate coefficient for the negative ions in the gas medium and higher degree of non-equilibrium in plasma, more instability will exist in the plasma.

In the following, we consider the oxygen plasma and calculate the relationship between the instability and certain factors (different electron temperatures, different attachment rate coefficients and different degrees of non-equilibrium in the plasma). Some rates coefficients of reactions in the oxygen plasma are listed in Table 1.[9−12]

Table 1. Rate coefficients of reactions in oxygen plasma with the electron temperature in units of eV and the heavy particle temperature in Kelvin.

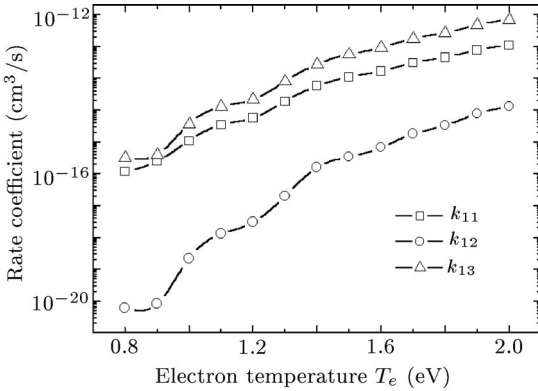|	Type	Reaction	Rate coefficient (cm^3/s)
1	Ionization	$e + O_2 \rightarrow O_2^+ + 2e$	$9.0 \times 10^{-10} T_e^{0.5} e^{(-12.6/T_e)}$
2	Ionization	$e + O_2 \rightarrow O + O^+ + 2e$	$5.3 \times 10^{-10} T_e^{0.9} e^{(-20/T_e)}$
3	Ionization	$e + O \rightarrow O^+ + 2e$	$9.0 \times 10^{-9} T_e^{0.7} e^{(-13.6/T_e)}$
4	Attachment	$e + O_2 \rightarrow O^- + O$	$8.8 \times 10^{-11} e^{(-4.4/T_e)}$
5	Attachment	$e + O_2 \rightarrow O^- + O^+ + e$	$7.1 \times 10^{-11} T_e^{0.5} e^{(-17/T_e)}$
6	Detachment	$O^- + O \rightarrow O_2 + e$	$6.1 \times 10^{-11} T_g^{1.05} e^{(-16998.6/T_g)}$

Fig. 1. Relationship between electron temperature T_e and rate coefficients of ionization reactions with k_{11}, k_{12} and k_{13} being the rate coefficients of ionization reactions 1–3 listed in Table 1.

Figures 1–4 show the calculation results for the oxygen plasma with the data listed in Table 1. Figure 1 displays the relationship between the electron temperature T_e and the rate coefficients of the ionization reactions, where k_{11}, k_{12} and k_{13} are the rate coefficients of ionization reactions 1–3, respectively. Figure 2 exhibits the relationship between the electron temperature T_e and the rate coefficients of attachment reactions, in which k_{21} and k_{22} are the rate coefficients

of attachment reactions 4 and 5 respectively. Figure 3 shows the relationship between the electron temperature and F for different rate coefficients of attachment reactions, where

$$F = \frac{k_i \hat{k}_i}{k_a \hat{k}_a} + \eta N \frac{k_d \hat{k}_d}{k_a \hat{k}_a}, \quad N = \frac{\bar{n}_n}{\bar{n}_e}. \tag{23}$$
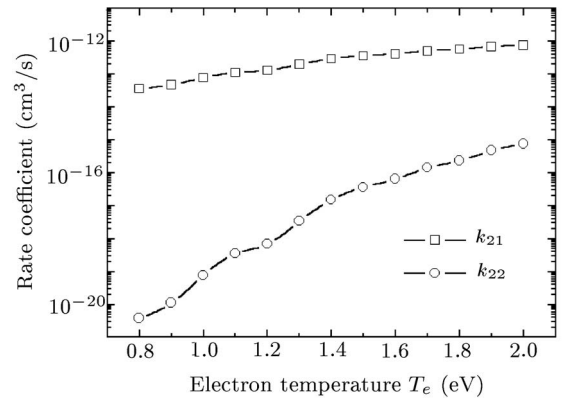

Fig. 2. Relationship between electron temperature T_e and rate coefficients of attachment reactions with k_{21} and k_{22} being the rate coefficients of attachment reactions 4 and 5.

It can be seen from Fig. 3 that when the medium gas has a greater attachment rate coefficient, there
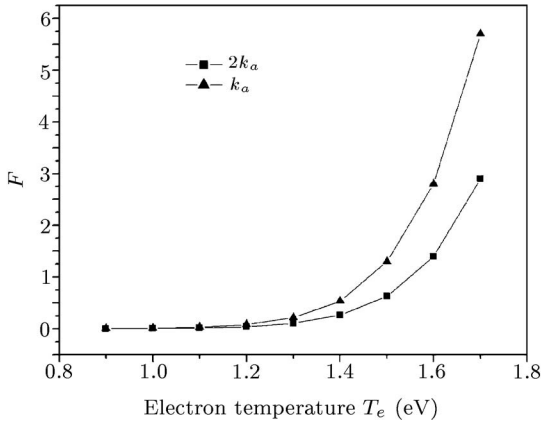
Fig. 3. Relationship between electron temperature and F for different rate coefficients of attachment reactions with $N = 0.8$ and $\eta = 0.1$.
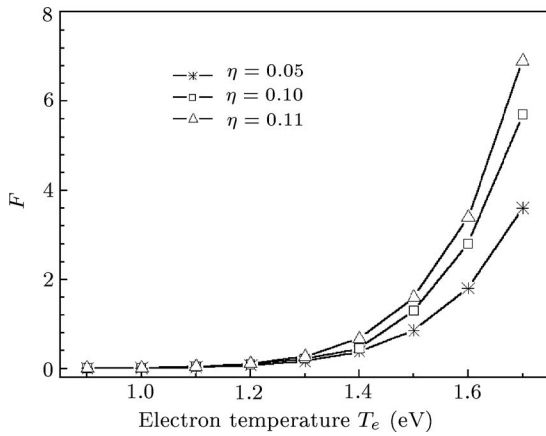

Fig. 4. Relationship between electron temperature and F for different η with $N = 0.8$.

appears to be greater instability in the plasma at the same electron temperature. Figure 4 shows the relationship between electron temperature and F for different η. It is obvious that the higher the degree of non-equilibrium, the greater the instability in plasma at the same electron temperature. Here η is related to the degree of non-equilibrium in plasma. When η is small, it means high degree of non-equilibrium in plasma.

In non-equilibrium plasma (including non-thermal plasma at atmospheric pressure and cold plasma at low pressure), there appear some instabilities with attachment processes of negative ions and degree of non-equilibrium in plasma. In Figs. 3 and 4, when $F < 1$, the plasma is unstable. In general, the greater the attachment rate coefficient and the higher the degree of non-equilibrium, the greater the instability in the plasma.

References

[1] Corr C S, Steen P G and Graham W G 2003 *Plasma Sources and Technology* **12** 265
[2] Lieberman M A, Lichtenberg A J and Marakhtanov A M 1999 *Appl. Phys. Lett.* **75** 3617
[3] Kunhardt E E 2000 *IEEE Trans. Plasma Sci.* **28** 189
[4] Stark R H and Schoenbach K H 1999 *J. Appl. Phys.* **85** 2075
[5] Haas R A 1973 *Phys. Rev.* A **8** 1017
[6] Nighan W L and Weigand W J 1974 *Phys. Rev.* A **10** 922
[7] Wang X Y and Cao J B 2003 *Chin. Phys. Lett.* **20** 1401
[8] Ma J Y, Qiu X J and Zhu Z Y 2003 *Chin. Phys. Lett.* **20** 1306
[9] Lieberman M A and Lichtenberg A J 1994 *Principles of Plasma Discharge and Materials Processing* (New York: Wiley) p 255
[10] Cresswell J D et al 1965 *NASA N 67-37521*
[11] Ma J and Pu Y K 2003 *Chin. Phys. Lett.* **20** 1527
[12] Kang Z D and Pu Y K 2002 *Chin. Phys. Lett.* **19** 1139