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Non-Linear Bistability of a Small Metallic Particle in Alternating Electric Field

L. G. Grechko, O. A. Davidova, V. M. Ogenko, and N. G. Shkoda

Institute of Surface Chemistry of NAS of Ukraine,
Ukraine, 03680, Kyiv-164, 17 General Naumov St.
E-mail: user@surfchem.freenet.kiev.ua

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The behavior of a small ellipsoidal metallic particle with non-linear (on the field) dielectric function in external alternating field is studied. In electrostatic approach the dependence of the linear enhancement factor of a local field on the particle's depolarization coefficient is calculated. The bistability conditions are found which appears under the account of a non-linearity, its nature and the boundaries of its existence are examined.

Calculation of the local electric field in small (compared to the wavelength of radiation) metallic or semiconductive particles with nonlinear dielectric function shows that the regimes exist where a given value of the external electric field may produce several different values of the local field and the polarization. This, in its turn, results in instability of the optical properties of the particles and of the disperse systems containing such particles. This phenomenon was called the intrinsic optical bistability (IOB) and has been intensively studied theoretically, being confirmed experimentally in connection with different applications [1-7].

The aim of the present study is a detailed theoretical analysis of enhancement of the local field as well as of the bistability domain (as intensity versus frequency of the incident electromagnetic wave) of particular metallic or semiconductive particle in the electrostatic approximation with account of cubic nonlinearity of its polarization with respect to the electric field.

In first section of the paper, we analyze the local field enhancement inside the ellipsoidal metallic particle embedded into a dielectric matrix in the case when the incident electric field is parallel to one of the ellipsoid axes. In second section, the condition of bistability, the bistability limits as well as the influence of the system parameters on these limits are studied in detail for such a particle.

We propose a comparatively simple analysis of roots of a cubic equation arising in a problem that may be useful for examination of the instability domains in the processes which are described by S-type characteristics.

1. Enhancement of Local Field in Small Metallic Ellipsoidal Particle

Let the electromagnetic wave falls on the metallic particle having a shape of rotational ellipsoid which is embedded into a dielectric host matrix The dielectric function (DF) of the particle depends on a frequency ω and the local electric field E (inside the particle) and may be presented in the form [1]

$$\varepsilon\left(\omega,\vec{E}\right)=\varepsilon(\omega)+\chi(\omega)\left|\vec{E}\right|^{2}, \qquad (1)$$

where $\chi(\omega)$ is the complex Kerr coefficient, $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$ is the linear part of DF and is taken in the Drude form

$$\varepsilon'(\omega)=\varepsilon_{\infty}-\frac{\omega_p^2}{\omega^2+\nu^2}, \quad \varepsilon''(\omega)=\frac{\nu}{\omega}\frac{\omega_p^2}{\omega^2+\nu^2}.$$

Here ω_p is the plasma frequency of electrons in the metal, ν is their collision frequency, ε_∞ is a constant that may be a function of frequency ω and depends on a particular metal.

Let the electric vector of the incident wave E_h be parallel to the large semi-axis of the ellipsoid. It is known [8] that in the electrostatic approximation (when the wave length of electromagnetic radiation is much larger than the particle typical size), the local field $\vec{E}$ is uniform and parallel to $\vec{E}_h$ for arbitrary dependence of $\varepsilon(\omega, \vec{E})$. It may be expressed in the form [8]

$$\vec{E} = F\vec{E}_h, \tag{2}$$

$$F = \frac{\varepsilon_h}{L}\frac{1}{\varepsilon' + \chi'\left|\vec{E}\right|^2 + i\left(\varepsilon'' + \chi''\left|\vec{E}\right|^2\right)},$$

where F is the enhancement factor, L is the depolarization factor along the field direction which in our case coincides with the larger semi-axis, ε_h is the dielectric function of a matrix, and ε' and ε^n are real and imaginary parts in the combination

$$\varepsilon \equiv \frac{\varepsilon_h(\omega)(1-L) + L\varepsilon(\omega)}{L}$$

χ' and χ'' are real and imaginary parts of $\chi(\omega)$, respectively.

Now we consider the case of such small electrical fields when $\chi = 0$. In this case Eq. (2) transforms into the expression

$$F_0 = \frac{\varepsilon_n}{L}\left[\left(\frac{1}{z_s^2} - \frac{1}{z^2+\gamma^2}\right) + \right.$$

$$\left. + i\left(\frac{1}{\overline{z}_s^2} + \frac{\gamma}{z\left(z^2+\gamma^2\right)}\right)\right]^{-1}. \tag{3}$$

Here we introduced dimensionless frequencies:

$$z = \frac{\omega}{\omega_p}, \quad \gamma = \frac{\nu}{\omega_p}, \quad z_s = \frac{\omega_s}{\omega_p}, \quad \overline{z}_s = \frac{\overline{\omega}_s}{\omega_p},$$

$$\omega_s = \omega_p\sqrt{\frac{L}{\varepsilon_h'(1-L) + \varepsilon_\infty' L}},$$

$$\overline{\omega}_s = \omega_p\sqrt{\frac{L}{\varepsilon_h''(1-L) + \varepsilon_s'' L}}.$$

It is worth noting that ω_s is the resonant plasma frequency of the metallic ellipsoid corresponding to direction of the electric field along the large semi-axis.

Now we analyze the dependence of $\left|F_0\right|^2$ on a frequency ω and the depolarization factor L. It is clear that the magnitude of $\left|F_0\right|^2$ considerably increases when the dimensionless frequency z approaches $z_0 = \left(z_s^2 - \gamma^2\right)^{1/2} \approx z_s$, i. e. the frequency of incident electromagnetic wave approaches the surface plasmon frequency (in the limit $\gamma \ll z_s$).

In particular, at $z = z_0$, Eq. (3) gives (when $\gamma^2 \ll z^2$)

$$\left|F_{0x}\right|^2 = \left|\frac{\varepsilon_h}{L}\right|^2 \left\{\gamma\left[\frac{\varepsilon_h}{L} + \varepsilon_\infty' - \varepsilon_h'\right]^{3/2} + \frac{\varepsilon_h''}{L} + \varepsilon_\infty'' - \varepsilon_h''\right\}^{-2}.$$

This function can have the extremum (maximum) by L in the interval [0, 1]. The extremum point $L = L_0$ can be found from the equation

$$(u+\alpha)^{1/2}(u - 2\alpha) = \delta, \tag{4}$$

where $\alpha = \dfrac{\varepsilon_\infty'}{\varepsilon_h'} - 1$, $\delta = \dfrac{\varepsilon_\infty'' - \varepsilon_h''}{\gamma(\varepsilon_h')^{3/2}}$, $u = \dfrac{1}{L}$.

For example, when $\delta \ll 0$ or $\gamma \gg \left|\varepsilon_\infty'' - \varepsilon_h''\right|$ the solution of Eq. (4) reads

$$L_0 = \frac{\varepsilon_h'}{2(\varepsilon_\infty' - \varepsilon_h')} \quad \text{or} \quad L_0 = \frac{\varepsilon_h'}{\varepsilon_h' - \varepsilon_\infty'}, \; \varepsilon_h' > \varepsilon_\infty'.$$

For finite δ, Eq. (4) can be solved numerically. If the point L_0 lies in the interval [0, 1], then $|F_{0s}|^2$ has the maximum in this interval at $L = L_0$ (Fig. 1). If L_0 is out of the specified interval then the larger $|F_{0s}|^2$ is realized at $L \to 1$. We note that Eq. (4) possesses only single real root.

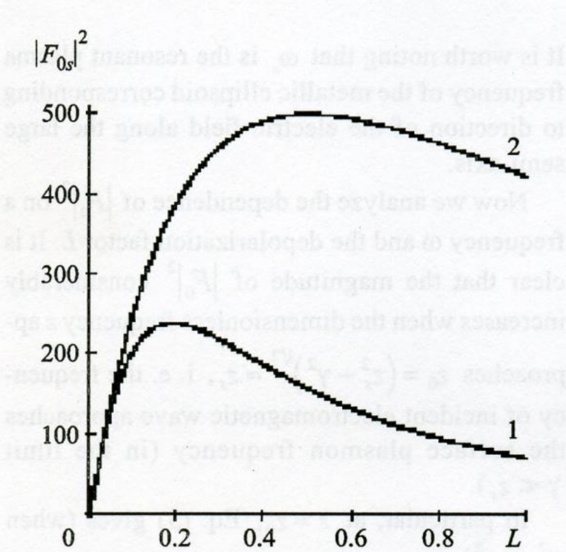


Fig. 1. *The enhancement factor $|F_{0s}|^2$ as a function of depolarization factor L:*
1 – a curve $|F_{0s}(L)|^2$ for a silver particle at $\varepsilon_\infty = 4.5 + i0.16$ and $\varepsilon_h = 2.25$ has a maximum at $L_0 = 0.189$;
2 – a curve $|F_{0s}(L)|^2$ for a silver particle at $\varepsilon_\infty = 4.5$ and $\varepsilon_h = 2.25$ has a maximum at $L_0 = 1/2$

2. Bistability in Ellipsoidal Metallic Particle with Nonlinear Dielectric Function

In this section we consider the local field in the metal ellipsoidal particle with account of the nonlinear part of $\varepsilon(\omega, \vec{E})$ in Eq. (1) which can be found from Eq. (2). Introducing the notations $|\chi||\vec{E}|^2 \equiv X$, $Y = \left|\frac{\varepsilon_h}{L}\right|^2 |\chi||\vec{E}_h|^2$ we obtain the following cubic equation for the variable X

$$X^3 + aX^2 + bX = Y, \qquad (5)$$

where

$$a = 2\left(\frac{\varepsilon'\chi' + \varepsilon''\chi''}{|\chi|}\right), \qquad b = |\varepsilon|^2.$$

This is the master equation for further analysis. It determines the dependence of the "local field" X on the "applied field" Y, frequency ω, and other parameters of the system that are hidden in coefficients a and b. The enhancement factor is given by a simple relation

$$|F|^2 = \left|\frac{\varepsilon_h}{L}\right|^2 \frac{X}{Y}.$$

Further, we will be interesting only in the real and positive roots of cubic Eq. (5). If this equation has one real positive root then the local field in the inclusion is a single-valued function of the applied field. If Eq. (5) has three positive roots then the local field is not a single-valued function of the applied field, and the system becomes unstable. Two ways of finding the root location of the cubic equation is described in Appendixes A and B.

Now we analyze the roots of Eq. (5) to find the IOB domain in the plane (*z*, *X*). According to Appendix A this equation has three real positive roots provided that conditions (A.8) hold true. In our case they may be written in the form

$$a \le -\sqrt{3b},$$

$$-\frac{2}{9}\left[Dx_2 + \frac{ab}{2}\right] \le Y \le -\frac{2}{9}\left[Dx_1 + \frac{ab}{2}\right], \qquad (6)$$

where

$$x_{1,2} = \frac{-a \mp \sqrt{D}}{3}, \qquad D = a^2 - 3b;$$

$$D = \frac{(\varepsilon'\chi' + \varepsilon''\chi'')^2 - 3(\varepsilon''\chi' + \varepsilon'\chi'')}{|\chi|^2}.$$

It follows from Eq. (6) that IOB occurs provided that $\varepsilon'' > 0$, $\chi'' > 0$ and quantities χ', ε' have different signs. Below, we carry out a detailed analysis of the case $\varepsilon' < 0$, $\chi' > 0$.

We consider an example of the non-absorbing host medium $(\varepsilon_h'' = 0)$ and the metal inclusion with the dielectric function $(\chi' > 0$ and $\chi'' = 0)$. In this case Eq. (5) takes the form

$$X^3 + 2\varepsilon' X^2 + |\varepsilon|^2 X = Y, \qquad (7)$$

where

$$\varepsilon'(z) = \frac{1}{z_s^2} - \frac{1}{z^2 + \gamma^2},$$

$$\varepsilon''(z) = \varepsilon''_\infty(z) + \frac{\gamma}{z(z^2 + \gamma^2)},$$

dimensionless frequencies z, z_s, γ are specified in the previous section. The first condition (6) can be written in the form

$$z^3\beta - z\left(z_s^2 - \gamma^2\beta\right) + \sqrt{3} z_s^2 \gamma \le 0, \qquad (8)$$

where $\beta = \left(1 + \sqrt{3} z_s^2 \varepsilon''_{s\infty}\right) > 0.$

To solve the inequality (8), we consider the cubic equation

$$z^3\beta - z\left(z_s^2 - \gamma^2\beta\right) + \sqrt{3} z_s^2 \gamma = 0. \qquad (9)$$

This equation according to Appendix B at $z_s^2 - \gamma^2\beta > 0$ has two positive roots z_2, z_3 and one negative root provided that its discriminant Q is negative, the coefficients of this equation are positive and lie close to the surface plasmon frequency $z^2 < z_s^2 - \gamma^2$. The boundary frequencies of IOB domain can be found from the cubic Eq. (7):

$$-(z_s^2 - \gamma^2\beta)^3 + \frac{81}{4} z_s^4 \gamma^2 \le 0.$$

As it follows from the Appendix A, at points z_2 and z_3, $a^2 - 3b = 0$ $(a = 2\varepsilon';\ b = |\varepsilon|^2)$. The critical magnitudes of electrical fields in these points are (the formula A.10)

$$(x_c)_{2,3} = -\frac{2}{3}\left(\frac{1}{z_s^2} - \frac{1}{z_{2,3}^2 + \gamma^2}\right)$$

$$(y_c)_{2,3} = (x_c)^3{}_{2,3}.$$

At $Q = 0$, $z_1 = z_2 = z_c$, the critical magnitude of field y_c, according to Eq. (9), coincides with the minimum value of the external electric field when the bistability occurs in the system. Therefore, in the case under consideration, the bistability in the system takes place provided that

$$z_2 \le z \le z_3;$$

$$-\frac{2}{9}\left(Dx_2 + \frac{ab}{2}\right) \le Y \le -\frac{2}{9}\left(Dx_1 + \frac{ab}{2}\right). \qquad (10)$$

Roots $z_2 > 0$, $z_3 > 0$ can be found from Eq. (8) which has three real roots.

We may note that from inequality (9) at $\gamma^2\beta / z_s^2 \ll 1$ one can get an approximate value of γ_c at fixed β and z_s .

$$\gamma_c = \frac{2}{9} z_s \left(1 - \frac{4}{81}\beta\right)^{3/2}.$$

If the collisional damping $\gamma > \gamma_c$ IOB disappears. We can assume that whatever increase in the damping of the metallic inclusion turns to the stiffer conditions of IOB. The minimal critical value of the external electric field of IOB origin can be found from Eq. (9). It is given by the relation

$$|\chi|\left|\vec{E}_h\right|_c^2 = \left(\frac{L}{\varepsilon_h}\right)^2 \left(\frac{1}{z_3^2+\gamma^2} - \frac{1}{z_s^2}\right)^3, \qquad (11)$$

where z_3 is the larger of roots z_2 and z_3 of Eq. (7). If $\varepsilon''_{s\infty}$ and γ tend to zero, the critical electric field tends to zero as well.

Figs. 2a, 3a show the dependences of $\chi\left|\vec{E}_s\right|^2$ on $\chi\left|\vec{E}_h\right|^2$ at different depolarization coefficients $L = 1/5$ (2a), and $L = 1/2$ (3a) at the frequency of electromagnetic field $\omega = 0.2\omega_p$. Figs. 2b, 3b show IOB domains in the plane "intensity" – dimensionless frequency of the electromagnetic wave $\left(\chi\left|\vec{E}_h\right|^2, z\right)$ (shadow area) which are determined from the relations (10) near the point z_3 (the larger root of Eq. (9)). The functions f_i are given by the expressions

$$f_i = -\frac{2}{9}\left(Dx_i + \frac{ab}{2}\right)\left(\frac{L}{\varepsilon_h}\right)^2,$$

$i = 1, 2.$

The limiting values of the incident electric field are shown in Figs. 2b, 3b by dash line at $\omega = 0.2\omega_p$. The whole bistability domain looks like an area enclosed into a hysteresis type curve. Its upper part gets narrowing with increasing the external field.

We would like to note that IOB in the spherical metal particle has been studied in [1]. But in this paper, only one root z_3 has been used. The entire domain of IOB was not discussed. More-

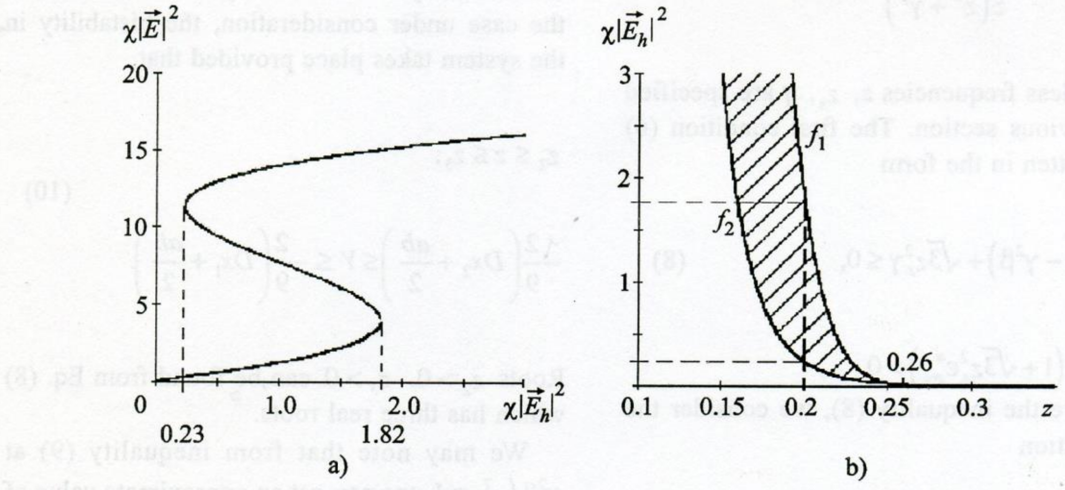


Fig. 2. *IOB in the ellipsoidal silver particle:* $\varepsilon'_\infty = 4.5$, $\varepsilon''_\infty = 0.16$, $\varpi_p = 1.46 \cdot 10^{16}\ s^{-1}$, $\nu = 1.68 \cdot 10^{14}\ s^{-1}$, $\varepsilon_h = 2.25$, $L = 0.2$ *(oblong spheroid* $z_2 = 0.16$, $z_3 = 0.26$*).*
a) dependence $\chi\left|\vec{E}\right|^2$ *on* $\chi\left|\vec{E}_h\right|^2$ *at* $z = z_2 = 0.16$*; b) IOB domain (shaded area) in the plane* $\left(\chi\left|\vec{E}_h\right|^2, z\right)$ *near the point* $z_3 = 0.26$ *(the larger positive root of Eq. (9))*

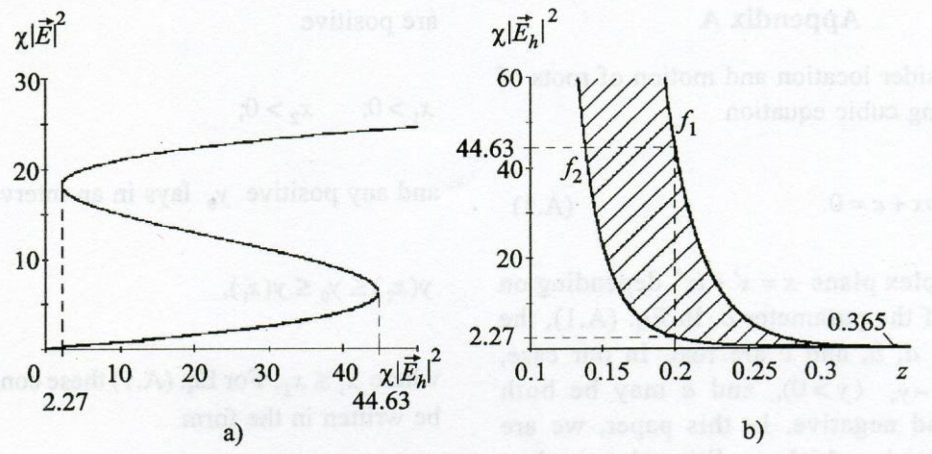


Fig. 3. *IOB in the ellipsoidal silver particle:* $\varepsilon'_\infty = 4.5$, $\varepsilon''_\infty = 0.16$, $\varpi_p = 1.46\cdot10^{16}\,s^{-1}$, $\nu = 1.68\cdot10^{14}\,s^{-1}$, $\varepsilon_h = 2.25$, $L = 0.5$ *(oblate spheroid* $z_2 = 0.023$, $z_3 = 0.365$*).*
a) Dependence $\chi\left|\vec{E}\right|^2$ *on* $\left(\chi\left|\vec{E}_h\right|^2, z\right)$ *at* $z = z_2 = 0.023$;
b) IOB domain (shaded area) in the plane $\left(\chi\left|\vec{E}_h\right|^2, z\right)$ *near the point* $z_3 = 0.365$ *(the larger positive root of Eq. (9))*

over, the approximation (9) made in the paper [1] does not allow one to specify the upper IOB boundary in the applied field.

Conclusion

The electrostatic response on external electric field of a small metallic particle with nonlinear DF displays some characteristic features. Even in a linear approximation ($\chi = 0$), under some definite values of the system parameters the extremum appears (at $L = 0.5$, see Fig. 1) on the plot of the enhancement factor $|F_0|^2$ versus L, for silver particles its value achieve 500. In the case $\chi \neq 0$ the bistability appears in the system caused by the possibility of existence of the S-like region on the plot of $I = \left|\vec{E}\right|^2$ versus $I_0 = \left|\vec{E}_h\right|$.

It should be noted that the appearance of such bistability in the problem under consideration ($\chi > 0$ and is real) is possible only when $\mathrm{Re}\,\varepsilon(\omega) < 0$, i. e. in frequency range of the existence of the surface plasmons in the ellipsoidal metallic particle. The bistability existence range is determined from the relation (10), and the minimum value of the external field $E_{0,c}$ at which it appears is found from Eq. (11). When the electron damping $\gamma = 0$, $E_{0,c} = 0$.

When γ increases, the bistability existence range by frequency becomes smaller, and at some value of γ it disappears. So, for the case shown in the Fig. 2 $\chi\left|\vec{E}_0\right|_c^2 = 0.23$. For metals $\chi \sim (2 \div 5)\cdot10^{-1}$ m^2/V [3]. From here, taking $\chi = 2.3\cdot10^{-11}$ m^2/V we find the estimate $E_{0,c} = 10^5$ V/m, i. e. the value which is completely achievable in the current laser systems. In this connection, the paper [3] should be noted where the bistability is experimentally examined in the composite including two-layered spherical particles (the core is a non-linear dielectric with the cubic non-linearity, and the shell is a metal), and its effect is studied on the composite scattering properties. The results obtained in our paper are of the principal interest for studying the scattering and absorption electromagnetic radiation by small particles in the frequency range where the bistability considered exists.

Appendix A

We consider location and motion of roots of the following cubic equation

$$x^3 + ax^2 + bx + c = 0. \qquad \text{(A.1)}$$

In the complex plane $x = x' + ix''$ depending on variation of the parameter c. In Eq. (A.1), the parameters a, b, and c are real. In our case, $b \geq 0$, $c = -y$, $(y > 0)$, and a may be both positive and negative. In this paper, we are interesting under which conditions imposed on the coefficients a, b, and y this equation has three (one) real positive roots. It is known that an answer to this question is given by the Routh – Hurwitz theorem [9]. The location of roots (A.1) depending on its coefficients that follows from the Routh – Hurwitz theorem is given in the Table of Appendix B. In particular, Eq. (A.1) has three real positive roots provided that

$$Q \leq 0, \quad b > 0, \quad y > 0, \quad ab + y > 0, \qquad \text{(A.2)}$$

where D is a discriminant of Eq. (A.1),

$$Q = \left(\frac{H}{3}\right)^3 + \left(\frac{q}{2}\right)^2;$$

$$H = \frac{a^3}{3} + b; \qquad \text{(A.3)}$$

$$q = 2\left(\frac{a}{3}\right)^3 - \frac{ab}{3} - y.$$

One can see that these conditions are rather complicated for the analysis. Here, we use a simplest way [5]. From the graphical analysis of Eq. (A.1) one can see that it has three (one) real roots if extremum points (if they exist) of the function

$$y = x^3 + ax^2 + bx, \qquad \text{(A.4)}$$

are positive

$$x_1 > 0; \qquad x_2 > 0; \qquad \text{(A.5)}$$

and any positive y_0 lays in an interval

$$y(x_2) \leq y_0 \leq y(x_1), \qquad \text{(A.6)}$$

where $x_1 \leq x_2$. For Eq. (A.1) these conditions can be written in the form

$$\begin{cases} a \leq -\sqrt{3b} \\ -\frac{2}{9}\left[(a^2 - 3b)x_2 + \frac{ab}{2}\right] \leq y_0 \leq -\frac{2}{9}\left[(a^2 - 3b)x_1 + \frac{ab}{2}\right]. \end{cases} \qquad \text{(A.8)}$$

Therefore, the intervals where cubic Eq. (A.1) has three real positive roots are given by the following expressions

$$\Delta(x) = x_2 - x_1 = \frac{2}{3}\left(a^2 - 3b\right)^{1/2},$$

$$\Delta(y) = y(x_1) - y(x_2) = \frac{4}{27}\left(a^2 - 3b\right)^{3/2} \qquad \text{(A.9)}$$

We may note that at $a^2 - 3b = 0$, $x_2 = x_1 = x_c$ and $y_2 = y_1 = y_c$, at the same time

$$x_c = \frac{a}{3}; \qquad y_c = \frac{a^3}{27}. \qquad \text{(A.10)}$$

The magnitudes x_c, y_c specify the critical values of x and y when three real positive roots appear in Eq. (A.1).

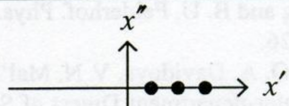
Appendix B

Range of parameters — ***Location of roots in a complex plane***

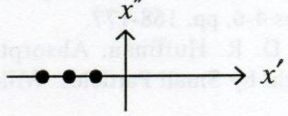
$x^3 + ax^2 + bx + c = 0,\qquad Q = (H/3)^3 + (G/2)^2 < 0,\qquad H = -a^2/3 + b,\qquad G = 2(a/3)^3 - ab/3 + c$

In this case all roots are real

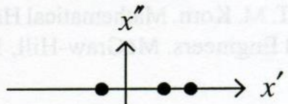
$ab - c < 0,$
$c < 0,\ b > 0$ (1)



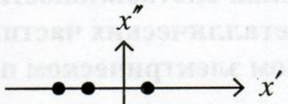
$ab - c < 0,$
$c > 0,\ b > 0$ (2)



$ab - c < 0,$
$c > 0,\ b >$ or (3)
$c > 0,\ b \le 0$



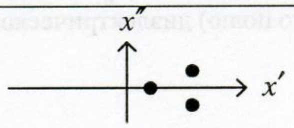
$ab - c < 0,$
$c < 0,\ b > 0$ or (4)
$c < 0,\ b \le 0$



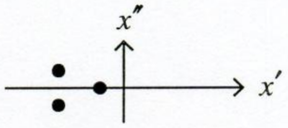
$x^3 + ax^2 + bx + c = 0,\qquad Q = (H/3)^3 + (G/2)^2 < 0,\qquad H = -a^2/3 + b,\qquad G = 2(a/3)^3 - ab/3 + c$

In this case one root is real and two roots are complex conjugated

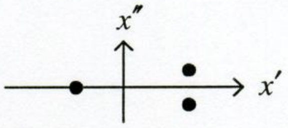
$ab - c < 0,$
$c < 0,\ b > 0$ (5)



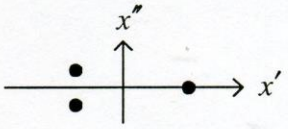
$ab - c < 0,$
$c > 0,\ b > 0$ (6)



$ab - c < 0,$
$c > 0,\ b > 0$ or (7)
$c > 0,\ b > 0$



$ab - c < 0,$
$c < 0,\ b > 0$ or (8)
$c < 0,\ b \le 0$



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Нелинейная бистабильность малых металлических частиц в переменном электрическом поле

Л. Г. Гречко, О. А. Давыдова, В. М. Огенко, Н. Г. Шкода

В работе изучено поведение малой металлической частицы эллипсоидальной формы во внешнем переменном электрическом поле с нелинейной (по полю) диэлектрической функцией. В электростатическом приближении рассчитана зависимость линейного фактора усиления локального поля от коэффициента деполяризации частицы. Изучен характер бистабильности, которая возникает в системе при учете нелинейности. Определены условия бистабильности и найдены границы ее существования.

Нелінійна бістабільність малих металевих частинок у змінному електричному полі

Л. Г. Гречко, О. А. Давидова, В. М. Огенко, Н. Г. Шкода

У роботі вивчено поведінку малої металевої частинки еліпсоїдальної форми у зовнішньому електричному полі з нелінійною (за полем) діелектричною функцією. У електростатичному наближенні розраховано залежність лінійного фактора підсилення локального поля від коефіцієнта деполяризації частинки. Вивчено характер бістабільності, яка виникає у системі внаслідок нелінійності. Знайдено умови бістабільності та визначено границі її існування.