POLARIZATION RELAXATION AND DETERMINISTIC CHAOS PHENOMENA IN DIELECTRIC SUSPENSION UNDER THE ACTION OF AC ELECTRIC FIELD

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1. Introduction. Pattern formation of colloidal systems under the action of electric field has caused a great interest recently [1, 2]. In particular the chevron like patterns of the stripes with circulating particles are observed in DNA solutions under the action of ac electric field [1]. An approach for the description of these patterns has been proposed in [3] where the polarization relaxation process of the cloud of contre ions surrounding DNA coils was considered. For the understanding of the different regimes of chevron band orientation in dependence on the frequency of the electric field the nonlinear polarization relaxation processes under the action of ac electric field should be investigated. However, as show recent investigations [4, 5], the polarization relaxation processes in dielectric suspensions can be very complicated. The deterministic chaos phenomena described by the polarization relaxation equations [6] was observed experimentally recently [7]. Here we investigate the regimes of polarization relaxation of dielectric suspension under the action of shear flow and ac electric field. Complex scenarious of the behaviour which include the period doubling transitions and deterministic chaos phenomena are found.

2. Model. Polarization relaxation equation is given by [6]

$$\frac{\mathrm{d}\mathbf{P}}{\mathrm{d}t} = [\mathbf{\Omega} \times \mathbf{P}] - \frac{1}{\tau}(\mathbf{P} - \chi\mathbf{E}), \tag{1}$$

here $\mathbf{\Omega}$ is the angular velocity of rotating particle, τ - the Maxwell relaxation time and $\chi = \chi_0 - \chi_\infty$, where χ_0 and χ_∞ are susceptibilities of particle polarization at low and high electric field frequencies respectively. Neglecting inertia of the small rotating particle, the balance of viscous and electrical torques reads

$$\alpha(\mathbf{\Omega} - \mathbf{\Omega}_0) = [\mathbf{P} \times \mathbf{E}], \tag{2}$$

here $\mathbf{\Omega}_0 = \Omega_0\mathbf{e}_z = const$ is the vorticity of macroscopic flow and α - the rotational friction coefficient of the particle. Introducing the frame, where $\mathbf{\Omega} = \Omega\mathbf{e}_z$, $\mathbf{E} = E\mathbf{e}_y \sin\omega t$ and $\mathbf{P} = P_x\mathbf{e}_x + P_y\mathbf{e}_y$, we get vectorial products $[\mathbf{P}\times\mathbf{E}] = EP_x\mathbf{e}_z \sin\omega t$ and $[\mathbf{\Omega} \times \mathbf{P}] = -\Omega P_y\mathbf{e}_x + \Omega P_x\mathbf{e}_y$. Thus by excluding Ω from equations (1,2) we obtain set of equations

$$\begin{cases} \dfrac{\mathrm{d}P_x}{\mathrm{d}t} = -\dfrac{E}{\alpha}P_xP_y\sin\omega t - \Omega_0 P_y - \dfrac{1}{\tau}P_x \\ \dfrac{\mathrm{d}P_y}{\mathrm{d}t} = (\dfrac{1}{\alpha}P_x^2 + \dfrac{\chi}{\tau})E\sin\omega t + \Omega_0 P_x - \dfrac{1}{\tau}P_y. \end{cases} \tag{3}$$

Introducing the period of ac field as the characteristic time scale and setting $P_i = \chi E \mathrm{P}_i$ in (3), dimensionless set of differential equations is obtained:

$$\begin{cases} \omega\dfrac{\mathrm{dP}_x}{\mathrm{dt}} = e\mathrm{P}_x\mathrm{P}_y\sin\mathrm{t} - \Omega_0\mathrm{P}_y - \mathrm{P}_x \\ \omega\dfrac{\mathrm{dP}_y}{\mathrm{dt}} = (1 - e\mathrm{P}_x^2)\sin\mathrm{t} + \Omega_0\mathrm{P}_x - \mathrm{P}_y, \end{cases} \tag{4}$$

where parameter e is expressed as follows: $e = E^2/E_c^2 = -\chi\tau E^2/\alpha$ but $\omega \to \omega\tau$ and $\Omega_0 \to \Omega_0\tau$ are given by ratio of the characteristic polarization relaxation time to the period of ac field and hydrodynamic orientation time respectively. It is known [6] that for dc external electrical field spontaneous rotation of particle occurs, when $E > E_c$, where $E_c^2 = -\alpha/\chi\tau$ (condition $\chi < 0$ is necessary).

Using scalar and pseudoscalar product, one can divide vector $\dot{\mathbf{P}}$ into parts parallel and orthogonal to $\mathbf{P}$, i.e., $\dot{\mathrm{P}}_\parallel = (\dot{\mathbf{P}} \cdot \mathbf{P})/|\mathbf{P}|$ and $\dot{\mathrm{P}}_\perp = (\dot{\mathbf{P}} \circ \mathbf{P})/|\mathbf{P}|$. From system (4) polar form equations can be obtained:

$$\begin{cases} \omega\dot{\mathrm{P}}_\parallel = \mathrm{z}\sin\varphi - \mathrm{P} \\ \omega\dot{\mathrm{P}}_\perp = \mathrm{z}(e\mathrm{P}^2 - 1)\cos\varphi - \Omega_0\mathrm{P}, \end{cases} \tag{5}$$

where φ is the angle with P_x axis and $\mathrm{z} = \sin\mathrm{t}$. From (5) one can see, that for any given value of variable z, all points satisfying $\dot{\mathrm{P}}_\parallel = 0$ lie on two circumferences with radius $\mathrm{z}/2$ and center $(0, \pm\mathrm{z}/2)$.

3. Stationary case. In the case, when $\omega \to 0$, the set of differential equations (4) reduces to algebraic set of equations

$$\begin{cases} e\mathrm{P}_x\mathrm{P}_y\mathrm{z} - \Omega_0\mathrm{P}_y - \mathrm{P}_x = 0 \\ (1 - e\mathrm{P}_x^2)\mathrm{z} + \Omega_0\mathrm{P}_x - \mathrm{P}_y = 0. \end{cases} \tag{6}$$

For given values of parameters e, Ω_0 and variable z, the first equation of (6) determines a hyperbola, but the second one - a parabola in $(\mathrm{P}_x, \mathrm{P}_y)$ plane. There will always be at least one common point for these two curves, but two common points (one crossing point and one touch point) or even three (all three crossing points) are also possible.

To classify these cases, we eliminate P_y from (6) and solve cubic equation for P_x. There will be a touching point, if two of found roots are equal. Thus we take arbitrary two roots $\mathrm{P}_{x,1}$ and $\mathrm{P}_{x,2}$ and set them to be equal. Equation $\mathrm{P}_{x,1} = \mathrm{P}_{x,2}$ gives three roots for z^2. Since we are interested in the region of (e, Ω_0) plane where $|\mathrm{z}| \leq 1$, we take an arbitrary root z_1^2 and by solving $\mathrm{z}_1^2 = 1$, we obtain the boundary of this region:

$$\Omega_c^2 = \frac{1}{8}\Big(\sqrt{e(e+8)^3} + e^2 - 20e - 8\Big), \tag{7}$$

where $e \geq 1$. If $\Omega_0^2 > \Omega_c^2$, then there is only one common point for hyperbola and parabola in each moment of time and imperfect bifurcation of the steady solutions of the polarization relaxation equations disappears. On the boundary, where $\Omega_0^2 = \Omega_c^2$, in addition to the one common point, periodical touching of curves occur. If $\Omega_0^2 < \Omega_c^2$, there is a periodic interchange between one and three common points, separated by momentary configurations of two points (touching of curves). For sufficiently large e, threshold value can be approximated as $|\Omega_c| \approx e/2$.

If we eliminate z from (6), we find that all crossing points of hyperbola and parabola must satisfy $\mathrm{P}_x\Big(e(\mathrm{P}_x^2 + \mathrm{P}_y^2) - 1\Big) = \Omega_0\mathrm{P}_y$ or in polar form:

$$\mathrm{P} = \sqrt{\frac{1 + \Omega_0\tan\varphi}{e}} \tag{8}$$

where φ is the angle with P_x axis. From (8) and (5) one can conclude, that in any given moment of time z, polarization vector $\mathbf{P}$ is located on the crossing point of curve (8) and circle $\mathrm{P} = \mathrm{z}\sin\varphi$. Thus for the case, when $\omega \to 0$, the trajectory of $\mathbf{P}$ coincides with the part of (8) being inside the circles with radius $1/2$ and centers $(0, \pm 1/2)$. Stationary trajectory $\mathbf{P} = 0$ is also possible.

4. Case without macroscopic rotation. If we assume, that there is no vorticity of macroscopic flow (i.e., $\Omega_0 \to 0$), system (4) reduces to

$$\begin{cases} \omega \dfrac{\mathrm{dP}_x}{\mathrm{dt}} = \mathrm{P}_x(e\mathrm{P}_y \sin \mathrm{t} - 1) \\ \omega \dfrac{\mathrm{dP}_y}{\mathrm{dt}} = (1 - e\mathrm{P}_x^2) \sin \mathrm{t} - \mathrm{P}_y. \end{cases} \tag{9}$$

In dependence on the parameters the solution with a nonzero transversal component of polarization averaged for the period of ac field exists. The range of parameters in which such solutions are possible can be estimated by asymptotics for small ω when the solution of the set (9) may be found in form ($\mathrm{P}_x = c(t_s); \mathrm{P}_y = a(t_s) \sin \mathrm{t} + b(t_s) \cos \mathrm{t}$) here $t_s = \omega \mathrm{t}$ is slow time of the system. As a result shortened set of equations is obtained

$$\begin{cases} \dfrac{\mathrm{d}a}{\mathrm{d}t_s} = -a + \omega b - c^2 e + 1 \\ \dfrac{\mathrm{d}b}{\mathrm{d}t_s} = -b - \omega a \\ \dfrac{\mathrm{d}c}{\mathrm{d}t_s} = \dfrac{1}{2} ace - c. \end{cases} \tag{10}$$

The stationary solution of set (10) shows that the supercritical bifurcation to the nontrivial solution $c = \pm\sqrt{e - 2(1 + \omega^2)/e}$ takes place at

$$e_c = 2(1 + \omega^2). \tag{11}$$

Thus for $e > e_c$ two symmetric periodic solutions with nonzero mean value of the transversal component of polarization exist. The phase portraits of these solutions are shown in Fig. 1 a). The numerical calculations show that the relation (11) for the critical electric field in dependence on the frequency holds unexpectedly well.

5. Chaos. The solutions with nonzero value of the mean transversal component of the polarization shown in Fig. 1*a* and *b* appear in some regions of the parameters for which the regular behaviour of the system takes place. They may be seen in bifurcation diagrams in Fig. 2*a* and *b* which are obtained by plotting the value of the polarization component after each period of the electric field. The period doubling scenario for the transition to the chaotic behaviour may be seen in Fig. 2*a* and *b*. Further understanding of the chaotic dynamics of the system may be obtained by construction of the flow of the points $(\mathrm{P}_x, \mathrm{P}_y)$ in the phase space and Poincare maps. Both show the characteristic "baker transformation" process (stretching and folding over).

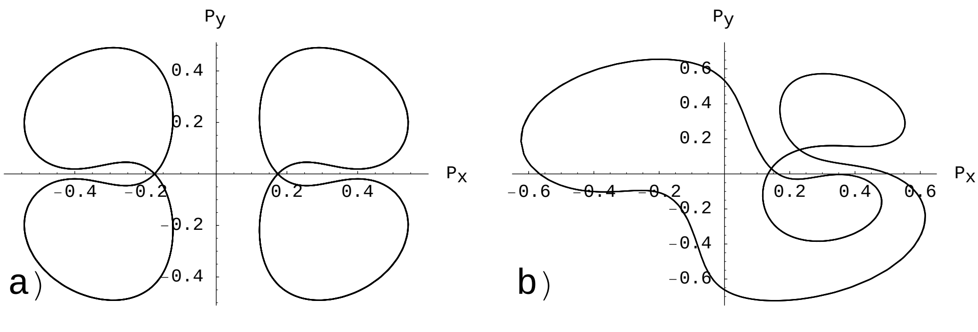

Fig. 1. Periodic trajectory of polarization vector **P**, where $e = 6.0$ in both cases and (*a*) $\Omega_0 = 0$, $\omega = 1.0$; (*b*) $\Omega_0 = 0.435$, $\omega = 1.0$.

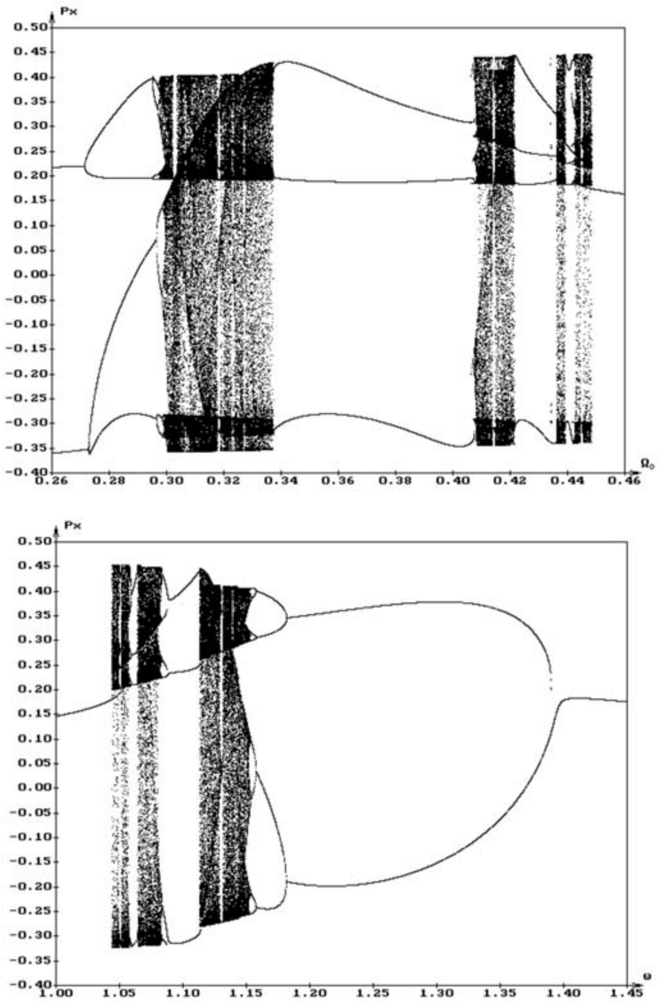

Fig. 2. (*a*) Bifurcation diagram of $P_x(\Omega_0)$, where $e = 6.0$ and $\omega = 1.0$, and (*b*) bifurcation diagram of $P_x(\omega)$, where $e = 6.0$ and $\Omega_0 = 0.5$ respectively.

6. Conclusions. This first study of the polarization relaxation of dielectric suspension under the action of ac electric field shows that the system has extraordinary rich behaviour. Several characteristic bifurcations of the system are described - non perfect bifurcation of the polarization equation solution in the case of the shear flow and the electric field of a low frequency, bifurcation to the periodic regime with nonzero mean value of the transversal component of the polarization, transition to the deterministic chaos phenomena by period doubling scenario and other.

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