Origin of the reduced attracting force between a rotating dielectric particle and a stationary one

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Recently Tao and Lan [Phys. Rev. E. **72**, 041508 (2005)] experimentally reported that the rotation of a dielectric particle can reduce significantly the attracting interparticle force between the rotating dielectric particle and a stationary one in argon gas. We develop the Gu-Yu-Hui theory of relaxation [J. Chem. Phys. **116**, 24 (2002)] to account for the Tao-Lan observations. Excellent agreement between the theoretical results and the Tao-Lan experimental data shows that the reduction in the attracting interparticle force is due to the effect of charge relaxation. We also show that the relaxation time of touching rotating particles can be accurately determined with the aid of the developed theory, for which, however, the well-known Maxwell-Wagner relaxation time is no longer valid.

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I. INTRODUCTION

The study of interparticle forces is important in many aspects of physics. For instance, if such forces are determined, the macroscopic structures of various physical systems may be determined. The interparticle forces between rotating particles are very important in the study of a variety of systems—e.g., colloidal suspensions including electrorheological fluids and magnetorheological suspensions, living cells suspensions, and so on—since the particles in these fluids may rotate under a shearing flow [1–10]. The study of the interparticle force between rotating dielectric particles has received much attention [11–14].

If two particles are stationary, polarization charges are caused to appear onto the surface of the particles in the presence of an external field. Once one of the particles starts to rotate, the polarization charges tend to deviate from the original position due to the rotation of the dielectric particle. On the other hand, the charges tend to return to the original position due to the directed external field. Thus, at dynamic equilibrium, the two competitive effects can be mediated by making the polarization charges locate in a position that differs from the original one, and thus the rotation changes the polarization charge distribution. As a result, this change affects the interaction between the two particles one of which is rotating.

Recently Tao and Lan [11] experimentally reported that the rotation of a dielectric particle can reduce significantly the attracting interparticle force between the rotating dielectric particle and a stationary one in argon gas. However, the physical mechanism behind the Tao-Lan observations has been up to now obscure [11]. To solve this, we shall present a relaxation theory, which is developed from the Gu-Yu-Hui theory [12] of relaxation. To this end, good agreement between the theoretical results and the Tao-Lan experimental data shows that the origin of the reduction in the attracting force between a rotating dielectric particle and a stationary one is due to a redistribution of the polarization charges on the surface of the rotating particle. Also, we shall show that the relaxation time of the touching rotating particle can be accurately determined by using the theory developed, for which, however, the well-known Maxwell-Wagner relaxation time is no longer valid.

The current paper is organized as follows. In Sec. II, we extend the Gu-Yu-Hui theory of relaxation to treat the polarization of higher orders being beyond the first order. This is followed by Sec. III where numerical results are presented and our results are compared to the Tao-Lan experiment. This paper ends with a discussion and conclusion in Sec. IV.

II. FORMALISM

According to the experiment setup designed by Tao and Lan [11], let us consider two spherical particles in an external electric field, one of which is held fixed and the other one rotates around the axis perpendicular to the line joining the centers of the two particles (Fig. 1). To proceed, we first briefly review the Gu-Yu-Hui theory of relaxation [12]. Let us consider two spherical dielectric particles without free charges arranged in a host medium—e.g., inertial gases like argon gas of dielectric constant 1.000 513. (The inertial gas can reduce the chance of dielectric breakdown and the electric field between the particles may not be strong enough to ionize the molecules.) There is an external applied electric field in the $\hat{\mathbf{z}}$ direction; see Fig. 1. According to the Gu-Yu-Hui theory of relaxation [12], the distribution $\sigma(\theta,\varphi,t)$ of the time-dependent surface-polarized charges on the rotating particle satisfies the relaxation equation

$$\frac{\partial\sigma(\theta,\varphi,t)}{\partial t}+\frac{\partial\sigma(\theta,\varphi,t)}{\partial\theta}\frac{\partial\theta}{\partial t}+\frac{\partial\sigma(\theta,\varphi,t)}{\partial\varphi}\frac{\partial\varphi}{\partial t}=-\frac{1}{\tau}[\sigma(\theta,\varphi,t)-\sigma_0(\theta,\varphi)], \quad (1)$$

where θ and φ are the polar angles in the spherical coordinates (r,θ,φ), with the origin being located at the center of the rotating particle, and τ is the relaxation time. By using the method originally introduced by Rayleigh [15], the surface charge distributions $\sigma_0(\theta')$ and $\sigma(\theta')$ of the stationary dielectric particle a and the rotating dielectric particle b are given as

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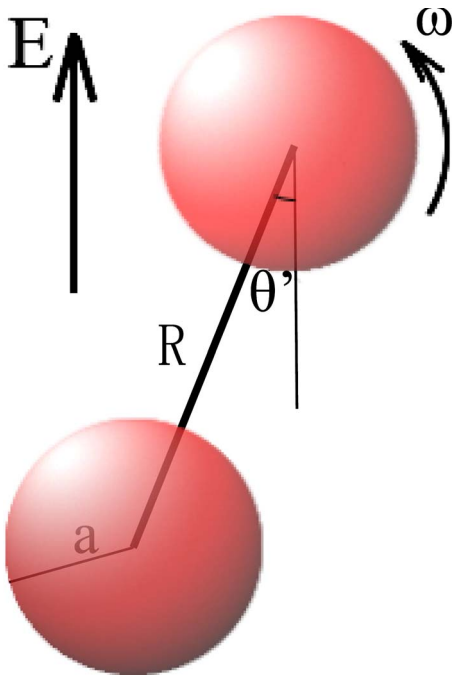

FIG. 1. (Color online) Schematic graph showing two spherical particles in an external electric field, one of which is held fixed and the other one rotates around the axis perpendicular to the line joining the centers of the two particles.

$$\sigma_0(\theta') = \sum_{l=1}^{\infty} H_l P_l(\cos\,\theta'), \tag{2}$$

$$\sigma(\theta') = \sum_{l=1}^{\infty} \widetilde{H}_l P_l(\cos\,\theta'), \tag{3}$$

where θ' is one polar angle in the spherical coordinates with the origin being located at the center of one particle, as shown in Fig. 1, P_l are the Legendre polynomials, and H_l and $\widetilde{H}_l$ are constants. To determine the induced attracting force between the stationary particle and the uniformly rotating particle, the electric potential may be evaluated in both the host and the rotating particle. And the energy of the system is

$$W = W_a + W_b = \frac{1}{8\pi}\sum_{\alpha=a,b}\int_{\Omega_\alpha}(\epsilon_h - \epsilon_\alpha)\mathbf{E}\cdot\mathbf{E}_0 d^3x, \tag{4}$$

where ϵ_h denotes the dielectric constant of the host and ϵ_α the dielectric constant of the particles a and b. In the following, for simplification, ϵ_α will be denoted as ϵ. At last, by taking the derivative of the energy, a formula relating the angular velocity of the rotating particle to the induced electric force on the particle is then obtained:

$$\mathbf{F} = \frac{1}{2}(1 + \zeta^{(1)})\mathbf{F}_0, \tag{5}$$

where $\mathbf{F}_0$ represents the interparticle force when both particles are at rest. Here only the lowest order of $\zeta^{(1)}[=1/[1+(\tau\omega)^2]]$ is given, which seems to be insufficient to interpret the Tao-Lan experiment data [11]. The data were shown to depend on the separation between the particles, which implies that the multipolar interaction between the particles should be taken into account. Thus, in this work, more perturbation terms are needed to interpret the Tao-Lan experiment results. After taking a lengthy calculation by taking more perturbation terms, we obtain the force between the rotating and the stationary particle analytically:

$$\mathbf{F} = \frac{1}{2}(1 + \zeta^{(1)} + \zeta^{(2)} + \cdots + \zeta^{(n)})\mathbf{F}_0, \tag{6}$$

where n stands for integers. To this end, our analysis shows that it is enough to keep terms up to third order, $\zeta^{(3)}$ [i.e., Eq. (7)], since the contribution of the other terms is weak enough to be neglected (see below). Then we obtain the interparticle force $\mathbf{F}$ as

$$\mathbf{F} = \frac{1}{2}(1 + \zeta^{(1)} + \zeta^{(2)} + \zeta^{(3)})\mathbf{F}_0, \tag{7}$$

where

$$\zeta^{(2)} = -\frac{15\pi T_2 a^4 R[20T_3(2P_4P_5 - 3P_3P_6)a^7 - 3P_3R^7]\tau\omega}{16[20T_2T_3a^{12}(5P_5^2 - 6P_4P_6) + 20T_3P_6a^7R^5 - 6T_2P_4a^5R^7 + R^{12}][1 + 4(\tau\omega)^2]}, \tag{8}$$

$$\zeta^{(3)} = \frac{14T_3a^5R^2[3T_2(4P_4^2 - 5P_3P_5)a^5 - 2P_4R^5](\tau\omega)^2}{[20T_2T_3a^{12}(5P_5^2 - 6P_4P_6) + 20T_3P_6a^7R^5 - 6T_2P_4a^5R^7 + R^{12}][1 + (\tau\omega)^2][1 + 9(\tau\omega)^2]}. \tag{9}$$

with a being the radius of the two particles. Here $P_l \equiv P_l(\cos\,\theta')$, $R(>2a)$ denotes the center-to-center separation between the two particles, and the coefficient T_l is given by

$$T_l = \frac{1 - \epsilon/\epsilon_h}{(1 + \epsilon/\epsilon_h) + 1/l}. \tag{10}$$

To comply with the experiments by Tao and Lan [11], let us set the line joining the centers of the two particles to be along the $\hat{\mathbf{z}}$ axis, and then $P_l(\cos\,\theta')$ turns out to be 1. Now $\zeta^{(2)}$ and $\zeta^{(3)}$ can be rewritten as

$$\zeta^{(2)} = \frac{15\pi T_2 a^4 R[20T_3 a^7 + 3R^7]\tau\omega}{16[-20T_2T_3a^{12} + 20T_3a^7R^5 - 6T_2a^5R^7 + R^{12}][1 + 4(\tau\omega)^2]}, \tag{11}$$

$$\zeta^{(3)} = -\frac{14T_3a^5R^2[3T_2a^5 + 2R^5](\tau\omega)^2}{[-20T_2T_3a^{12} + 20T_3a^7R^5 - 6T_2a^5R^7 + R^{12}][1 + (\tau\omega)^2][1 + 9(\tau\omega)^2]}. \tag{12}$$

From Eqs. (11) and (12), it is evident that the reduction of the interparticle attracting force [Eq. (7)] depends on the gap between the two particles.

III. NUMERICAL RESULTS

In Fig. 2, we investigate the reduction of the attracting force F/F_0 versus the separation R between the centers of the two particles for the perturbation terms up to first order $[\mathbf{F}=(1/2)(1+\zeta^{(1)})\mathbf{F}_0]$, second order $\left[\mathbf{F}=\frac{1}{2}(1+\zeta^{(1)}+\zeta^{(2)\prime})\mathbf{F}_0\right]$ with $\zeta^{(2)\prime}=(45\pi T_2a^4RP_3\tau\omega)/\{(16[-6T_2P_4a^5+R^5][1+4(\tau\omega)^2])\}$, and third order $[\mathbf{F}=(1/2)(1+\zeta^{(1)}+\zeta^{(2)}+\zeta^{(3)})\mathbf{F}_0]$. We find that the addition of the high-order terms plays a significant role in the reduction of the attracting force, especially as the gap between two particles is small (in this case the multipolar interaction between the particles becomes more evident). Nevertheless, if the two particles separate in distance, the first-order approximate is good enough already.

From Fig. 2 [or Eq. (5)], we can see that the force up to the first order $\zeta^{(1)}$ (which is already showed in Ref. [12]) is unrelated to the separation R, because it is within the dipole approximation. In this situation, the relation between the interparticle force and the separation is contained only in the function of $\mathbf{F}_0$. The force up to the second order $\zeta^{(2)\prime}$ is within the quadrupole approximation, while that up to the third order $\zeta^{(3)}$ is within the octopole approximation. When the two particles are separated far away, the full expression $\mathbf{F}$ [Eq. (6)] reduces to Eq. (5). That is, in this case the two particles can be seen as point dipoles. This is both physical and reasonable. In addition, we have also calculated $\zeta^{(n)}$ with n up to 30 numerically and found that the result predicted by the expressions with terms up to nth order ($n>3$) is similar to that up to $n=3$ only. So, in this work, the neglect of the terms of higher orders is reasonable as well [Eq. (7)].

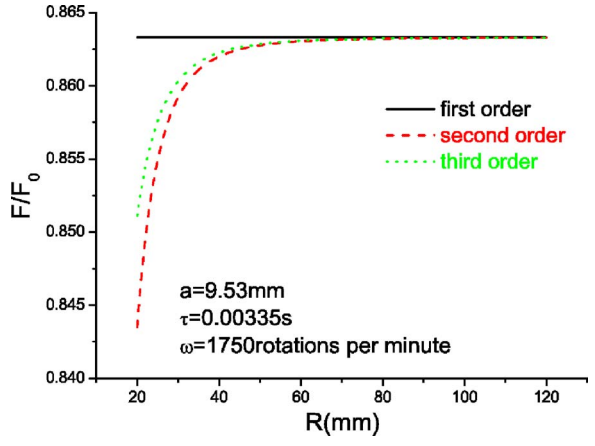

FIG. 2. (Color online) The effect of the terms up to first order $[\mathbf{F}=\frac{1}{2}(1+\zeta^{(1)})\mathbf{F}_0]$, second order $\left[\mathbf{F}=\frac{1}{2}(1+\zeta^{(1)}+\zeta^{(2)\prime})\mathbf{F}_0\right]$ with $\zeta^{(2)\prime}=(45\pi T_2a^4RP_3\tau\omega)/\{(16[-6T_2P_4a^5+R^5][1+4(\tau\omega)^2])\}$, and third order $\left[\mathbf{F}=\frac{1}{2}(1+\zeta^{(1)}+\zeta^{(2)}+\zeta^{(3)})\mathbf{F}_0\right]$ on the reduced interparticles force F/F_0, versus the separation R between the centers of a stationary dielectric particle and a rotating one. Parameters: $\epsilon_1=2.5\epsilon_0$ and $\epsilon_2=1.000513$.

Figure 3 shows the reduction of the attracting force versus rotation frequency ω (in unit of rotations per minute) with the modification of the high-order terms. It is apparent to see that increasing ω leads to decreasing interparticle force $\mathbf{F}$. In the meantime, the high-order terms can offer a correction.

Figure 4 displays a fitting of Tao and Lan's experimental data of a rotating dielectric particle (made of polyamide of dielectric constant 2.5) and a stationary dielectric one (made of the same polyamide of dielectric constant 2.5) [11] by using the presently developed theory [Eq. (7)]. For the fitting, we first fit the case of $R=19.314$ mm (i.e., gap $=0.381$ mm) and $E=100$ V/m by choosing an appropriate relaxation time $\tau=0.003\,347$ s, according to which the other two groups of experimental data are fitted very well. Here it should be remarked that the relaxation time $\tau=0.003\,347$ s is reasonably comparable to that of particles suspended in electrorheological fluids [16]. According to the numerical results, we find that the relaxation time of the rotating dielectric particle is about 10^{-3} s. However, for the stationary dielectric sphere, Maxwell-Garnett theory [17] can be used to predict the well-known Maxwell-Wagner relaxation time (see Appendixes A and B) $\tau_{MW}=(\epsilon+2\epsilon_h)/(\sigma+2\sigma_h)$, where σ and $\sigma_h$mean the conductivity of the particle and the host argon gas, respectively. From the experiments by Tao and Lan [11],

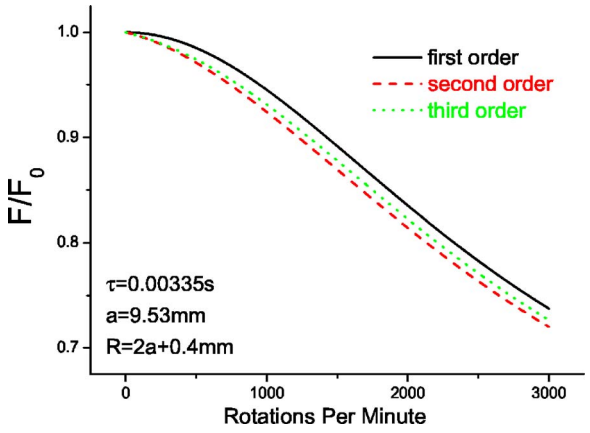

FIG. 3. (Color online) Same as Fig. 2, but versus the rotating frequency of the particle.

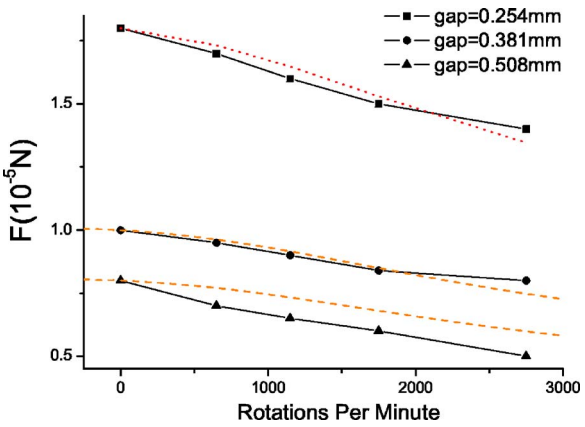

FIG. 4. (Color online) Fitting of the experimental data of a rotating dielectric particle and a stationary dielectric one in an electric field. The symbols denote the data which are extracted from Ref. [11]. According to our theory [Eq. (7)], the dotted line is plotted by choosing an appropriate relaxation time τ=0.003 347 s, for the fitting of the case of R=19.314 mm (i.e., gap=0.381 mm). The relaxation time τ=0.003347 s is used for the fitting of the other two cases. Good agreement between the theory and the experiment is shown.

we have $\sigma=10^{-10}$ S/m and $\sigma_h \approx 0$, thus yielding τ_{MW}=0.3983 s. In this case, we can conclude that the expression for the Maxwell-Wagner relaxation time τ_{MW} does not hold for touching rotating dielectric spherical particles. And the leaky dielectrics process which the Maxwell-Wagner relaxation time [17] comes from (see Appendixes A and B) may not be the exact source of the relaxation time of the touching rotating particles. Generally speaking, the surface property of touching rotating dielectric particles is likely to bring such a short relaxation time $\sim 10^{-3}$ s, due to the effect of multipolar interaction. For displaying the theoretical results in Fig. 4, we have assumed that the relaxation time of the touching rotating particle is unchanged, even if the separation between the two particles is changed. In fact, the interaction between the particles also has an effect on the relaxation time, too. To determine the relation between relaxation time τ and separation R, we try to use Eq. (7) to fit each group of data extracted from the Tao-Lan experiments of a rotating dielectric particle and a stationary one. For obtaining an excellent agreement between the data and the developed theory [Eq. (7)], more accurate relaxation times τ should be used instead, as listed in Table I. It is shown that the relaxation times of the touching rotating dielectric spherical particles relate to their separation and thus the multipolar interaction and that they take the values with the same order of magnitude for the current systems. The latter implies the reason why the three groups of experimental data are fitted very well by using a single relaxation time τ=0.003 347 s.

IV. DISCUSSION AND CONCLUSION

Here some comments are in order. The Gu-Yu-Hui theory of relaxation [12] is valid for dielectric-dielectric, metallic-metallic, and dielectric-metallic particles under dc electric fields and for dielectric-dielectric particles under ac electric fields. In the Tao-Lan experiments [11], they used ac fields at frequency 300 Hz. Thus, in this work we have focused on dielectric-dielectric particles only. As shown above, the theory can help to determine the relaxation time of touching rotating dielectric particles, for which the usual Maxwell-Wagner relaxation time (see Appendixes A and B) is no longer valid to predict the accurate value. The reason is that the usual Maxwell-Wagner relaxation time holds only for isolated (or nontouching) stationary particles [18]. In this work, while one τ is extracted from one group of experimental data, it can be used to fit the other two groups of data very well. This shows that the τ was determined accurately in this way. However, this τ is two orders of magnitude smaller than that evaluated from the Maxwell-Wagner relaxation. This shows that the Maxwell-Wagner relaxation seems to be incorrect here. For the case of touching rotating particles (rather than that of isolated stationary particles), one might estimate the relaxation time by comparing experimental data (e.g., those by Tao and Lan [11]) and the current theory. On the other hand, for treating touching rotating particles, we suggest that the Maxwell-Wagner relaxation be improved by including the effects of dynamics and multipolar interaction arising from touching particles.

TABLE I. Relaxation time τ which should be used to achieve a good agreement between the present theory [Eq. (7)] and the Tao-Lan experimental data of a rotating dielectric sphere (polyamide) and a stationary dielectric one (polyamide). For comparison, the Maxwell-Wagner relaxation time τ_{MW} is also calculated.

F_{st} (10^{-5} N)	E_0 (V/mm)	R (mm)	τ (10^{-3} s)	τ_{MW} (10^{-1} s)
1.0	100	19.441	3.093	3.983
1.8	100	19.314	3.347	3.983
0.8	100	19.568	5.838	3.983

In Appendixes A and B, we have derived the Maxwell-Wagner relaxation time for either an infinite large plank or a dielectric sphere by solving the charge conservation law and the corresponding boundary conditions. Based on this, it is apparent that the Maxwell-Wagner relaxation time is only determined by the dielectric constant and electric conductivity of the plank or sphere. For our system of two touching particles, one of which is rotating, both the dynamic effects and the interparticle interaction are expected to contribute together to the surprisingly short relaxation time that differs from the relaxation time predicted by the Maxwell-Wagner theory.

To sum up, we have developed the Gu-Yu-Hui theory of relaxation to account for the Tao-Lan observations of reduced attracting forces between a rotating dielectric particle and a stationary one. We have shown that the origin of the reduction in the attracting forces is due to a redistribution of the polarization charges on the surface of the dielectric particle rotating around its center. Also, the relaxation time of the rotating particle can be accurately extracted by comparing the theory with the experimental data, for which the well-known Maxwell-Wagner relaxation time fails. This work is of value for the study of dynamic electrorheological fluids or

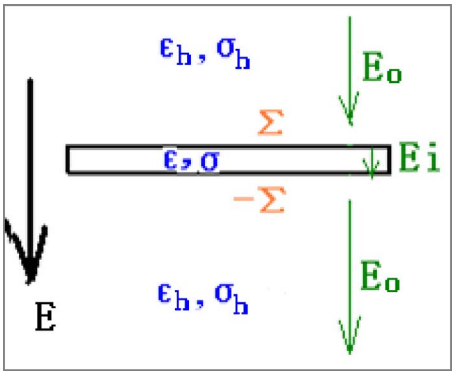

FIG. 5. (Color online) Sketch of an infinite large plank in an applied electric field $E_0\hat{\mathbf{z}}$.

other kinds of colloidal suspensions, or even living cells suspensions.

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APPENDIX A: THE MAXWELL-WAGNER RELAXATION TIME FOR AN INFINITE LARGE PLANK

Let us consider an infinite large plank in an applied electric field $E_0\hat{\mathbf{z}}$; see Fig. 5. The dielectric constant of the plank and the host medium is ϵ and ϵ_h, while the conductivity is σ and σ_h, respectively. The electric field inside the plank is $\mathbf{E}_i$, and on the edge of the plank, the electric field outside is $\mathbf{E}_h$. When the field is suddenly applied, the charge in the plank has not been redistributed. So we have $E_h=E_0$ and $E_i=E_0$ at first. After a long time, the whole system come to its stationary state. With the theory of electrodynamics, we have $E_h=E_0$ and $E_i=\frac{\epsilon_h}{\epsilon}E_0$ in that condition. Further assuming that the relation between the electric field outside E_h and that inside E_i is linear, just as

$$\mathbf{E}_f\cdot\vec{n}=a\mathbf{E}_i\cdot\vec{n}+b\mathbf{E}_0\cdot\vec{n}, \tag{A1}$$

we obtain

$$\mathbf{E}_h\cdot\vec{n}=\mathbf{E}_i\cdot\vec{n}+(E_0-\mathbf{E}_i\cdot\vec{n})=E_0. \tag{A2}$$

The appropriate boundary conditions are

$$\Sigma|_{bou}=\epsilon_h\mathbf{E}_h\cdot\vec{n}-\epsilon\mathbf{E}_i\cdot\vec{n}, \tag{A3}$$

$$\nabla\cdot\mathbf{j}|_{bou}=\sigma_h\mathbf{E}_h\cdot\vec{n}-\sigma\mathbf{E}_i\cdot\vec{n}. \tag{A4}$$

At last, we consider the charge conservation law

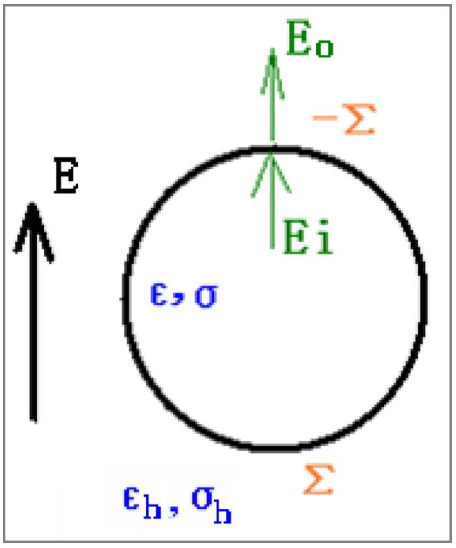

FIG. 6. (Color online) Sketch of a dielectric sphere in an applied electric field $E_0\hat{\mathbf{z}}$.

$$\frac{\partial\Sigma}{\partial t}+\nabla\cdot\mathbf{j}=0. \tag{A5}$$

Solving Eqs. (A2)–(A5), we obtain

$$\Sigma(t)\sim(1-e^{-t/\tau}), \tag{A6}$$

where the relaxation time τ is

$$\tau=\frac{\epsilon}{\sigma}. \tag{A7}$$

APPENDIX B: THE MAXWELL-WAGNER RELAXATION TIME FOR A DIELECTRIC SPHERE

If a sphere (Fig. 6) takes the place of the plank we discussed in Appendix A, we can calculate the relaxation time in the same way. When the field is suddenly applied, the system has $E_h=E_0$ and $E_i=E_0$, which is the same as the plank case. After the whole system comes to its stationary state, there may be a free charge $\Sigma\cos(\theta)$ on the surface. On the polar coordinates of the sphere (where $\theta=0$), we have $E_h=\frac{3\epsilon}{\epsilon+2\epsilon_h}E_0$ and $E_i=\frac{3\epsilon_h}{\epsilon+2\epsilon_h}E_0$ in that case. Further using Eq. (A1) we obtain

$$\mathbf{E}_h\cdot\vec{n}=\mathbf{E}_i\cdot\vec{n}+3(E_0\cos\theta-\mathbf{E}_i\cdot\vec{n})=3E_0\cos\theta-2\mathbf{E}_i\cdot\vec{n}. \tag{B1}$$

Solving Eqs. (A3)–(A5) and (B1), we obtain

$$\Sigma(t)\sim(1-e^{-t/\tau}), \tag{B2}$$

where the relaxation time τ is

$$\tau=\frac{\epsilon+2\epsilon_h}{\sigma+2\sigma_h}. \tag{B3}$$

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