Alternating-current relaxation of a rotating metallic particle*

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Based on a first-principles approach, we establish an alternating-current (AC) relaxation theory for a rotating metallic particle with complex dielectric constant $\varepsilon_\alpha = \bar{\varepsilon}_\alpha - \mathrm{i}\bar{\sigma}_\alpha/\omega_0$. Here $\bar{\varepsilon}_\alpha$ is the real part, $\bar{\sigma}_\alpha$ the conductivity, ω_0 the angular frequency of an AC electric field, and $\mathrm{i} = \sqrt{-1}$. Our theory yields an accurate interparticle force, which is in good agreement with the existing experiment. The agreement helps to show that the relaxations of two kinds of charges, namely, surface polarized charges (described by $\bar{\varepsilon}_\alpha$) and free charges (corresponding to $\bar{\sigma}_\alpha$), contribute to the unusually large reduction in the attracting interparticle force. This theory can be adopted to determine the relaxation time of dynamic particles in various fields.

Keywords: AC relaxation theory, rotating metallic particle, interparticle force

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1. Introduction

The structures of various systems, such as colloidal suspensions including electrorheological fluids,[1–3] magnetorheological suspensions,[4–7] ferrofluids,[8–11] living cells suspensions,[12] and so on, are very sensitive to the change of interparticle forces. Unfortunately, some shearing flow that is inevitable in experiments and natural environments can cause the rotation of the colloidal particles,[13–23] thus reducing the interparticle force to some extent. An interaction between a rotating electric field and suspended particles or cells can also lead to a rotational motion of the particles. This phenomenon, known as electrorotation, has been increasingly employed as a sensitive tool for noninvasive studies of a broad variety of microparticles, ranging from living cells to nanowires, seeds as well as synthetic materials.[24–29] In addition, insulating particles suspended in a liquid with low conductivity can also rotate in the presence of an electric field, which leads to the so-called negative effective viscosity effect.[30,31]

The dielectric relaxation can affect the inter-particle force, and even the property of the semi-conducting materials.[32–34] So far, the relaxation of polarized charges in dielectric particles has been investigated within the well-known Maxwell–Wagner relaxation theory. However, it was shown that the Maxwell–Wagner relaxation time is valid for an isolated stationary dielectric sphere or infinite large plank, whose dielectric responses are represented by a dielectric constant and electric responses by a conductivity.[35]

When a dielectric particle 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 change the polarization charge distribution. As a result, this change affects the interaction between the two particles one of which is rotating. The same process also happens when a metallic particle substitutes for the dielectric one. However, for the extremely high conductivity of the metallic material, it is believed that free charges on the surface of the metallic particle will not cause an obvious force decrease in a low rotation speed. On the contrary, Tao and Lan[36] experimentally reported that the rotation of a metallic particle can reduce significantly the attracting interparticle force between the rotating metallic particle and a stationary dielectric or metallic one in argon gas. To the best of our knowledge, this was the first direct experiment conducted on the interparticle force between two touching rotating particles. Tan *et al.*[37] found that the local electric field between two metallic particles weakens as one rotates faster, which can be used to qualitatively explain the phenomenon of Tao and Lan's work.[36] However, a quantitative explanation is still lacking for explaining Tao and Lan's observations,[36] which is important for practice. To solve this, we believe that the relaxation processes in dynamic metallic particles should be understood carefully, which are also expected to receive a broad interest in various fields related to dynamics in (soft) condensed matter. Reference [38] described a method to solve similar systems.

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However, only dielectric particles were under their consideration. In this work, we improve the method in Ref. [38]. Based on a first-principles approach, we establish a relaxation theory for dynamic metallic particles in the presence of an external AC field, thus being called AC relaxation theory.

This paper is organized as follows. In Section 2, we establish an AC relaxation theory for dynamic metallic particles on the basis of a first-principles approach. In Section 3, the comparison between theory and experiment is presented, which is followed by a conclusion in Section 4.

2. Formalism

2.1. Static charge distribution

Let us consider two spherical particles in an external electric field $E_0\widehat{\boldsymbol{Z}}$, one of which is held fixed. The other one rotates around the axis perpendicular to the line joining the centers of the two particles with the rotation frequency, ω, see Fig. 1.

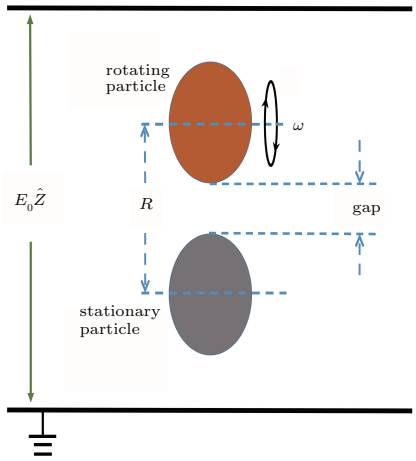

Fig. 1. (color online) Schematic graph showing two spherical particles in an external AC electric field, $E_0\widehat{\boldsymbol{Z}}$, one of which is held fixed and the other of which rotates around the axis perpendicular to the line joining the centers of the two particles with the rotation frequency, ω. R represents the center-to-center distance between the two particles, and "gap" denotes the surface-to-surface separation between the two particles.

The value of ω can be controlled artificially (e.g., the rotating particle may be connected to a motor[36]). In spherical coordinates, the electric potential Φ satisfies the Laplace equation and the general expressions for $\Phi_h(\boldsymbol{r}_\alpha)$ in the host region in the vicinity of particle α can be written down as[39]

$$\Phi_h(\boldsymbol{r}_\alpha) = A_0^\alpha + \sum_{l=1}^{\infty}\left(A_l^\alpha r_\alpha^l + \frac{B_l^\alpha}{r_\alpha^{l+1}}\right)\mathrm{P}_l(\cos\theta_\alpha),\ \alpha = a,b, \tag{1}$$

and the potential inside particle α, $\Phi_i(\boldsymbol{r}_\alpha)$, is[39]

$$\Phi_i(\boldsymbol{r}_\alpha) = C_0^\alpha + \sum_{l=1}^{\infty} C_l^\alpha r_\alpha^l \mathrm{P}_l(\cos\theta_\alpha),\quad \alpha = a,b, \tag{2}$$

where P_l are the Legendre polynomials. There is no coordinate component φ, for we simply assume the rotating symmetry. The coefficients A_l^α, B_l^α, and C_l^α are yet to be determined.

Applying the boundary conditions, i.e., the continuity of the potential and the electric displacement on the surface of particles, we obtain two sets of relations between the unknown coefficients:

$$A_l^\alpha = \frac{1}{T_l^\alpha + 1} C_l^\alpha, \tag{3}$$

$$B_l^\alpha = \frac{a_\alpha^{2l+1}}{1/T_l^\alpha + 1} C_l^\alpha, \tag{4}$$

where

$$T_l^\alpha = \frac{1-\varepsilon_\alpha/\varepsilon_h}{(1+\varepsilon_\alpha/\varepsilon_h)+1/l},\quad \alpha = a,b. \tag{5}$$

Here ε_α and ε_h are the dielectric constants of particle α and the host, respectively, and a_α is the radius of particle α.

Since we try to introduce the effect of free charges which may exist on the surface of metallic particles, the conductivity of the particle should be considered. In that case, the complex dielectric constant ε_α can be written as

$$\varepsilon_\alpha = \bar{\varepsilon}_\alpha - \mathrm{i}\bar{\sigma}_\alpha/\omega_0, \tag{6}$$

where $\bar{\varepsilon}_\alpha$ is the real part of the complex dielectric constant, $\bar{\sigma}_\alpha$ the conductivity of the metallic particle, ω_0 the angular frequency of the applied AC electric field, and $\mathrm{i} = \sqrt{-1}$. Consequently, the potential $\Phi_i(\boldsymbol{r})$ inside a particle consists of a real part $\Phi_{R,i}(\boldsymbol{r})$ and an imaginary part $\Phi_{I,i}(\boldsymbol{r})$ as

$$\Phi_i(\boldsymbol{r}) = \Phi_{R,i}(\boldsymbol{r}) + \mathrm{i}\,\Phi_{I,i}(\boldsymbol{r}). \tag{7}$$

The polarized and free charge distributions σ_{st} on the surface of the metallic particle are related to the discontinuity in the electric field across the surface through

$$\sigma_{R,\mathrm{st}}(\theta_\alpha,\varphi_\alpha) = \left(\frac{\partial\Phi_{R,i}(\boldsymbol{r})}{\partial r} - \frac{\partial\Phi_{R,h}(\boldsymbol{r})}{\partial r}\right)\Bigg|_{r=a_\alpha},$$
$$\sigma_{I,\mathrm{st}}(\theta_\alpha,\varphi_\alpha) = \left(\frac{\partial\Phi_{I,i}(\boldsymbol{r})}{\partial r} - \frac{\partial\Phi_{I,h}(\boldsymbol{r})}{\partial r}\right)\Bigg|_{r=a_\alpha}. \tag{8}$$

In the calculation below, we deal with the complex parameters directly.

The remaining unknown coefficients C_l^α can be found by constructing a set of Rayleigh identities by the Green function formulation. According to the Green function approach,[39] the effects of the polarized charges on the potential at any location can be written as

$$\Phi(\boldsymbol{r}_\alpha) = E_0 z_\alpha + \frac{1}{4\pi}\sum_{\beta=a,b}\int q_\beta(\boldsymbol{R})G(\boldsymbol{r}_\beta - \boldsymbol{R})\,\mathrm{d}\boldsymbol{R}. \tag{9}$$

Here $\boldsymbol{r}_\beta$ is a position vector originated from the center of particle b to the same location as $\boldsymbol{r}_\alpha$. The parameter $q(\boldsymbol{R})$ represents the surface polarized charge, which is obtained from Eq. (8) in our system.

Comparing Eqs. (1), (2), and (9), which represent two different ways of describing the same quantity in regions occupied by the particles, we obtain a pair of Rayleigh identities given by

$$\sum_{l=1}^{\infty} A_l^{\alpha} r_{\alpha}^{l} \mathrm{P}_l(\cos\theta_{\alpha}) = -E_0 z_{\alpha} + \sum_{l=1}^{\infty} \frac{B_l^{\beta}}{r_{\beta}^{l+1}} \mathrm{P}_l(\cos\theta_{\beta}), \quad \alpha = a, b. \tag{10}$$

In the experiment of Ref. [36], the two particles were arranged along the $\hat{Z}$ direction. To comply with this, in the current work we also set the applied electric field to be $\hat{Z}$-directed (Fig. 1). Consider the origin at the center of particle α, i.e., $r_{\alpha} = 0$, and $r_{\beta} = R$ (here R is the distance between the centers of the two particles). Taking the partial derivatives of Eq. (10) with respect to r, we obtain a set of linear equations for the unknown coefficients

$$A_n^{\alpha} = -E_0 \delta_{n,1} + \sum_{l=1}^{\infty} (-)^n \frac{(n+l)! B_l^{\beta}}{n! l! R^{n+l+1}}, \quad \alpha = a, b. \tag{11}$$

Thus, all the coefficients A_l^{α}, B_l^{α}, and C_l^{α} in the potential are determined. Please refer to Ref. [38] for more detailed derivation. We can get the first-order parameters

$$C_1^{\alpha} = \frac{3R^3 \varepsilon_h E_0 \cos(\omega_0 t)}{2a_{\alpha}^3(\varepsilon_{\alpha} - \varepsilon_h) - R^3(\varepsilon_{\alpha} + 2\varepsilon_h)}. \tag{12}$$

Here ω_0 is the frequency of the applied field. For particles with different dielectric constant and radius, the coefficients C_1^a and C_1^b are different. Furthermore, we can write the surface charge of a stationary particle as

$$\sigma_{\mathrm{st}}(\theta_{\alpha}) = \sum_{l=1}^{\infty} H_l \mathrm{P}_l(\cos\theta_{\alpha}), \tag{13}$$

where the first-order term is

$$H_1 = \frac{3R^3(\varepsilon_{\alpha} - \varepsilon_h) E_0 \cos(\omega_0 t)}{2a_{\alpha}^3(\varepsilon_{\alpha} - \varepsilon_h) - R^3(\varepsilon_{\alpha} + 2\varepsilon_h)}. \tag{14}$$

The parameters are complex forms as described by Eq. (6).

2.2. Charge distribution on rotating particles

Once the rotation appears, the displaced original polarized and free charge distributions deviate from the static distribution. The distribution will tend to relax to an equilibrium distribution at a certain rate $1/\tau$ where τ means the relaxation time, which is dependent on the material forming the particles, the geometry of the particles, and some other elements like rotation speed and circumstance. Here, according to Eq. (6), the relaxation time τ can be separated as τ_R and τ_I. The former (τ_R) results from $\bar{\varepsilon}_{\alpha}$ and the latter (τ_I) is due to $\bar{\sigma}_{\alpha}$. That is, they are related to surface-polarized charges (σ_R) and free charges (σ_I), respectively. The time-dependent $\sigma_R(\theta,\varphi,t)$ and $\sigma_I(\theta,\varphi,t)$ on a rotating metallic particle satisfy the following two equations

$$\begin{aligned}
&\frac{\partial \sigma_R(\theta,\varphi,t)}{\partial t} + \frac{\partial \sigma_R(\theta,\varphi,t)}{\partial \theta}\frac{\partial \theta}{\partial t} + \frac{\partial \sigma_R(\theta,\varphi,t)}{\partial \varphi}\frac{\partial \varphi}{\partial t} = -\frac{1}{\tau_R}[\sigma_R(\theta,\varphi,t) - \sigma_{R,\mathrm{st}}(\theta,\varphi)],\\
&\frac{\partial \sigma_I(\theta,\varphi,t)}{\partial t} + \frac{\partial \sigma_I(\theta,\varphi,t)}{\partial \theta}\frac{\partial \theta}{\partial t} + \frac{\partial \sigma_I(\theta,\varphi,t)}{\partial \varphi}\frac{\partial \varphi}{\partial t} = -\frac{1}{\tau_I}[\sigma_I(\theta,\varphi,t) - \sigma_{I,\mathrm{st}}(\theta,\varphi)],
\end{aligned} \tag{15}$$

where t is time, and θ or φ is the polar angle in the spherical coordinates (r,θ,φ) with the origin being located at the center of the rotating particle. In our system, the axis of rotation is perpendicular to the applied field and the line joining the centers of the particles (Fig. 1), and the charge re-distribution due to rotation possesses symmetry break, yielding the dependence of charge distribution on φ. However, its correction to interaction forces was found to be small enough to be neglected, according to the method presented in Ref. [40]. Thus, in what follows, the dependence on φ will be dropped. We can use coefficients $\widetilde{H}_l$ to expand the charge distribution $\sigma(\theta)$ on the rotating particle in terms of Legendre polynomials

$$\sigma(\theta_{\alpha}) = \sum_{l=1}^{\infty} \widetilde{H}_l \mathrm{P}_l(\cos\theta_{\alpha}), \tag{16}$$

where $\widetilde{H}_l$ are related to the other coefficients we calculated in Subsection 2.1. By obtaining the surface charge distributions, we are able to achieve the potential inside or outside particles, when one of them is rotating.

2.3. Force exerted on rotating particles

The force exerted on a particle can be found by evaluating the derivative of the energy with respect to particle displacement in space. The change in electrostatic energy due to the introduction of particles into the host medium in the presence of an electric field $\boldsymbol{E}_0$ is[39]

$$\begin{aligned}
W &= \frac{1}{16\pi}\int [(\boldsymbol{E}^* \cdot \boldsymbol{D}_h + \boldsymbol{E} \cdot \boldsymbol{D}_h^*) - (\boldsymbol{D}^* \cdot \boldsymbol{E}_0 + \boldsymbol{D} \cdot \boldsymbol{E}_0^*)]\mathrm{d}^3 x\\
&= \frac{1}{8\pi}\sum_{\alpha=a,b}\int_{\Omega_{\alpha}} \mathrm{Re}\,[(\varepsilon_h - \varepsilon_{\alpha})\boldsymbol{E}] \cdot \boldsymbol{E}_0 \mathrm{d}^3 x \equiv W_a + W_b,
\end{aligned} \tag{17}$$

where ε_{α} stands for the complex dielectric constant of particle a and b, respectively. Here, $\mathrm{Re}\,[\cdots]$ denotes the real part of $\cdots$, $\boldsymbol{D}$ represents the electric displacement, and the local field $\boldsymbol{E}$ is given by $\boldsymbol{E} = -\nabla\Phi$. Let $W_{\mathrm{st},\alpha}$ be the change in energy due to the presence of particle α but the particle is kept

stationary. The first-order energy of a stationary particle is

$$W_{\mathrm{st},\alpha} = -\frac{V}{8\pi}\mathrm{Re}\left[(\varepsilon_h - \varepsilon_\alpha)C_1^\alpha\right]E_0, \tag{18}$$

where V is the volume of the particle under consideration. The comparison between W_α and $W_{\mathrm{st},\alpha}$ leads to the following relation

$$W_\alpha = \zeta W_{\mathrm{st},\alpha}, \tag{19}$$

where ζ is a coefficient varying in different systems.

Now, by taking the derivative of the energy, a formula relating the angular velocity ω of the rotating particle to the induced force $\boldsymbol{F}$ on the particle is then obtained as

$$\boldsymbol{F} = \frac{\partial W_{\mathrm{st},a}/\partial R + \zeta \partial W_{\mathrm{st},b}/\partial R}{\partial W_{\mathrm{st},a}/\partial R + \partial W_{\mathrm{st},b}/\partial R}\boldsymbol{F}_{\mathrm{st}}, \tag{20}$$

where $\boldsymbol{F}_{\mathrm{st}}$ represents the interparticle force when both particles are at rest. By performing calculations, the exact value of the coefficient ζ is found to be

$$\begin{aligned}\zeta &= \frac{\omega_0(\varepsilon_h - \bar{\varepsilon}_\alpha)\mathrm{Re}\left[\widetilde{H}_1\right] - \bar{\sigma}_\alpha \mathrm{Im}\left[\widetilde{H}_1\right]}{\omega_0(\varepsilon_h - \bar{\varepsilon}_\alpha)\mathrm{Re}\left[H_1\right] - \bar{\sigma}_\alpha \mathrm{Im}\left[H_1\right]} \\ &= \frac{\omega_0(\varepsilon_h - \bar{\varepsilon}_\alpha)\left[2J_R\sum_{n=1}^{\infty} G_e(2n)\mathrm{Re}\left[H_{2n}\right] - 3\sum_{n=0}^{\infty} V_R(2n+1)\mathrm{Re}\left[H_{2n+1}\right]\right]}{\omega_0(\varepsilon_h - \bar{\varepsilon}_\alpha)\mathrm{Re}\left[H_1\right] - \bar{\sigma}_\alpha \mathrm{Im}\left[H_1\right]} \\ &\quad - \frac{\bar{\sigma}_\alpha\left[2J_I\sum_{n=1}^{\infty} G_e(2n)\mathrm{Im}\left[H_{2n}\right] - 3\sum_{n=0}^{\infty} V_I(2n+1)\mathrm{Im}\left[H_{2n+1}\right]\right]}{\omega_0(\varepsilon_h - \bar{\varepsilon}_\alpha)\mathrm{Re}\left[H_1\right] - \bar{\sigma}_\alpha \mathrm{Im}\left[H_1\right]}.\end{aligned} \tag{21}$$

Here, we only keep the first three terms in our numerical calculation for each relaxation time, and the contributions from all the other terms are small enough to be neglected. The coefficients V_R, V_I, J_R, J_I, and G_e are respectively given by

$$V_R(2n+1) = \sum_{k=0}^{n}\frac{\lambda^c(2k+1)G_o(2n+1,k)}{1+(2k+1)^2\tau_R{}^2\omega^2}, \tag{22}$$

$$V_I(2n+1) = \sum_{k=0}^{n}\frac{\lambda^c(2k+1)G_o(2n+1,k)}{1+(2k+1)^2\tau_I{}^2\omega^2}, \tag{23}$$

$$J_R = \frac{3\pi\tau_R\omega}{8(1+4\tau_R^2\omega^2)}, \tag{24}$$

$$J_I = \frac{3\pi\tau_I\omega}{8(1+4\tau_I^2\omega^2)}, \tag{25}$$

$$G_e(n) = \frac{(n-3)!!\,(n+1)!!}{2^{n-1}\,(n/2-1)!\,(n/2+1)!}, \tag{26}$$

where

$$G_o(n,k) = \frac{(n-2k-2)!!(n+2k)!!}{2^{n-1}\left((n-1)/2-k\right)!\left((n-1)/2+k+1\right)!}, \tag{27}$$

$$\lambda^c(k) = -\frac{2^{k-1}\,k!}{(2k+1)!!}\prod_{j=1}^{k/2}\frac{k^2-(k-2j+3)^2}{k^2-(k-2j)^2}. \tag{28}$$

In Eqs. (13) and (21), the constants H_l can be calculated by the method in Subsection 2.1. In these equations, ζ is related to the rotation frequency ω. When both particles are at rest, we have $\zeta = 1$. The rotation causes ζ to have different values, thus yielding $F \neq F_{\mathrm{st}}$.

3. Comparison between theory and experiment

In the experiment,[36] two spherical particles of diameter 19.06 mm were arranged vertically inside a capacitor composed of two horizontal electrodes. One of the particles, which could move up and down to adjust the gap between particles, was connected to a motor and was able to rotate around the horizontal axis. Another one rested on a microbalance. The whole device was placed inside a glove box, which was filled with dry argon gas. When there was no electric field, the reading on the microbalance was just the weight of the stationary particle. When an electric field was applied, the induced attracting force between the rotating metallic particle (cooper) and the stationary metallic particle (cooper) or the dielectric one (polyamide) reduced the reading on the microbalance. Thus, the attracting force between the two particles was able to be determined.

Now we are in a position to compare theory and experiment. For this purpose, consistent with the experiment,[36] the diameters of particles are also set to be 19.06 mm. The distance between the centers of the two particles, R, which is used in our calculations, can be got by $R = \mathrm{gap} + 19.06$ mm, where the gap is the surface-to-surface separation between the particles (see Fig. 1). The values of gap are acquired from the experiment.[36] By using Eq. (20), we obtain a fitting of Tao and Lan's experimental data of a rotating metallic particle (made of copper of dielectric constant 6) and a stationary metallic one (Fig. 2) or a stationary dielectric one (Fig. 3).[36] For the fitting, we first fit the case of $R = 19.441$ mm (or gap$= 0.381$ mm) by choosing appropriate relaxation times, $\tau_R = 4.342\times10^{-4}$ s and $\tau_I = 7.3\times10^{-8}$ s for the metallic–metallic system (Fig. 2) and $\tau_R = 4.352\times10^{-4}$ s and $\tau_I = 3.6\times10^{-11}$ s for the metallic–dielectric system (Fig. 3), according to which the other groups of experimental data are also fitted well. As ω increases, the attracting force between the rotating particle and the stationary one decreases. However, such a reduction of the force weakens with gap enlarging.[36] Here it should be remarked that the relaxation time τ_R used for this fitting is reasonably comparable to that of the particles

suspended in electrorheological fluids.[41]

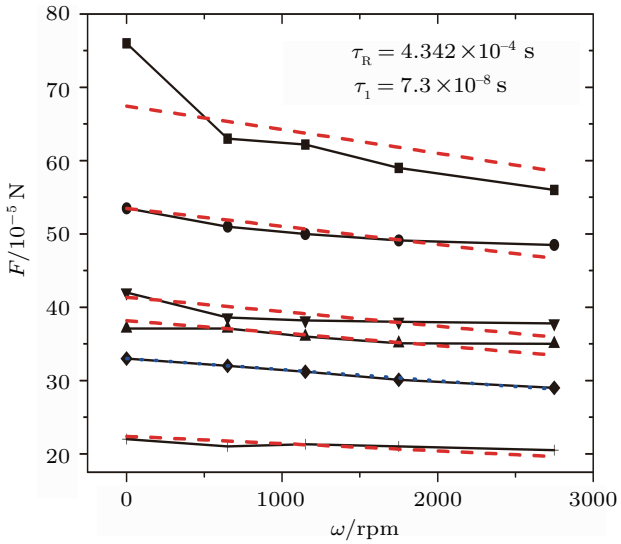

Fig. 2. (color online) Fitting of the experimental data of a rotating metallic particle (copper) and a stationary metallic one (copper) in an AC electric field with frequency $\omega_0 = 300$ Hz. The symbols denote the experimental data which are extracted from Ref. [36]. From upper to bottom: $R = 19.314$ mm (or gap =0.254 mm) and $E_0 = 100$ V/mm; $R = 19.441$ mm (or gap =0.381 mm) and $E_0 = 100$ V/mm; $R = 19.314$ mm (or gap =0.254 mm) and $E_0 = 80$ V/mm; $R = 19.568$ mm (or gap =0.508 mm) and $E_0 = 100$ V/mm; $R = 19.441$ mm (or gap =0.381 mm) and $E_0 = 80$ V/mm; $R = 19.568$ mm (or gap =0.508 mm) and $E_0 = 80$ V/mm. Similarly, according to our theory [Eq. (20)], the dotted line is plotted by choosing appropriate relaxation times, $\tau_R = 4.342\times10^{-4}$ s and $\tau_I = 7.3\times10^{-8}$ s, for the fitting of the case of $R = 19.441$ mm (or gap$= 0.381$ mm) and $E_0 = 80$ V/mm. The two values, $\tau_R = 4.342\times10^{-4}$ s and $\tau_I = 7.3\times10^{-8}$ s, are then used for fitting the other five groups of the experimental data (dashed lines). In this figure, the unit of ω, "rpm", stands for "rotations per minute".

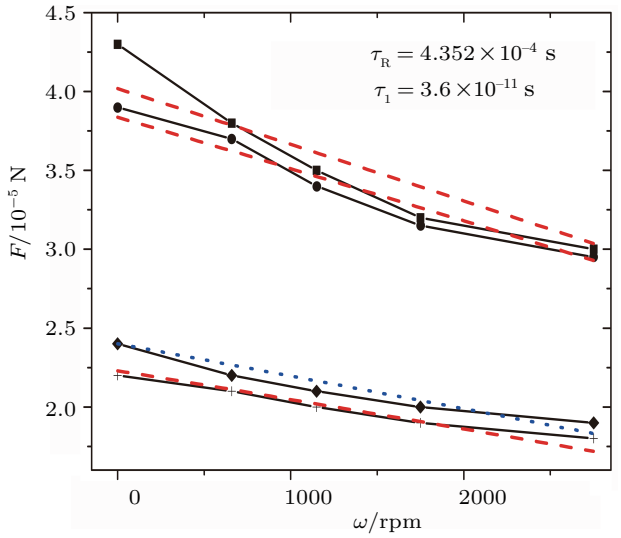

Fig. 3. (color online) Fitting of the experimental data of a rotating metallic particle (copper) and a stationary dielectric one (polyamide) in an AC electric field with frequency $\omega_0 = 300$ Hz. The symbols denote the experimental data which are extracted from Ref. [36]. From upper to bottom: $R = 19.314$ mm (or gap =0.254 mm) and $E_0 = 100$ V/mm; $R = 19.441$ mm (or gap =0.381 mm) and $E_0 = 100$ V/mm; $R = 19.441$ mm (or gap =0.381 mm) and $E_0 = 80$ V/mm; $R = 19.568$ mm (or gap =0.508 mm) and $E_0 = 80$ V/mm. According to our theory [Eq. (20)], the dotted line is plotted by choosing appropriate relaxation times, $\tau_R = 4.352\times10^{-4}$ s and $\tau_I = 3.6\times10^{-11}$ s, for the fitting of the case of $R = 19.441$ mm (or gap$= 0.381$ mm) and $E_0 = 80$ V/mm. The two relaxation times, $\tau_R = 4.352\times10^{-4}$ s and $\tau_I = 3.6\times10^{-11}$ s, are then used for fitting the other three groups of the experimental data (dashed lines).

For the rotating metallic sphere, the well-known Maxwell–Wagner relaxation time[35,42] predicts $\tau_{\rm MW} = (\bar{\varepsilon}_\alpha + 2\varepsilon_h)/(\bar{\sigma}_\alpha + 2\sigma_h)$, where ε_h (or σ_h) means the real part of the complex dielectric constant (or conductivity) of the host argon gas. According to the experiments by Tao and Lan,[36] we have $\bar{\sigma}_\alpha = 5.8\times10^7$ S/m and $\sigma_h \approx 0$, thus yielding $\tau_{\rm MW} = 1.22\times10^{-18}$ s, which is much smaller than both of the above-mentioned fitting parameters, τ_R and τ_I. It is worth noting that charge imbalances could relax in a period of time, which is comparable to the characteristic collision time (τ') of electrons in conductors,[43] $\tau' = \bar{\sigma}_\alpha m/ne^2$. Here m, n, and e are the electron mass, average electron density, and electron charge, respectively. It is because the time dependence is so rapid that the inertia of electrons must be taken into account. For copper, one obtained $\tau' \approx 2\times10^{-14}$ s.[43] In view of the precision of the experiments reported in Ref. [36], we expect that the collision time (τ') should be comparable to the relaxation time τ_I. Then we can conclude that neither the collision process described by τ' nor the leaky dielectrics process characterized by $\tau_{\rm MW}$[35,42] serves as the source of the relaxation time τ_R.

According to our results, the relaxation time τ_R is generally much longer than τ_I. In fact, the τ_R sources from polarized charges (described by $\bar{\varepsilon}_\alpha$) under the local field of touching particles, as mentioned in Subsection 2.2. Because of the short separation between the particles, a strong local field is formed, and shifts the property of electrons in metallic particles. When a particle is rotating, the local field should change greatly.[37] Thus, the interaction force decreases.[36] In general, the change of local field of touching rotating metallic particles is likely to bring such a long relaxation time $\sim 10^{-4}$ s, due to the effect of multipolar interactions. As far as we know, theories of this process based on the first principles have not yet been found for predicting τ_R directly. In this situation, our method is a possible way to obtain τ_R of metallic particles by comparing with experiment data. On the other hand, the relaxation time τ_I, which originates from free charges (corresponding to $\bar{\sigma}_\alpha$), is much shorter. The collision time as mentioned above can be used to understand the general vanishing value of τ_I. We can conclude that τ_I refers to the inertia of electrons in conductors. Since τ_R is much longer, we believe that the dielectric relaxation process plays a key role in the force reduction in the experiment.

4. Discussion and conclusion

When displaying the theoretical results in Figs. 2 and 3, we have assumed that the relaxation times of the touching rotating particle are unchanged, even if the separation between the two particles or the external electric field is changed. In fact, these factors also have an effect on the relaxation times, τ_R and τ_I. Therefore, we try to use Eq. (20) to fit the experimental data, by using more accurate relaxation times, as listed

in Tables 1 and 2 explicitly. It is found that the relaxation times τ_R always take the values with the same order of magnitude for the current systems. The stability of the values of τ_R is an evidence of the validity of our fitting presented in Figs. 2 and 3. For the relaxation time τ_I ranges over several orders of magnitude, we think that either the limit of experimental precision or the inaccuracy of the fitting process is a possible reason. As mentioned above, the relaxation time τ_I is vanishing (compared with τ_R). Thus, tiny fluctuations of experimental data may lead to a great change of the fitting values of τ_I.

Table 1. Relaxation times, τ_R and τ_I, for obtaining good fitting with experimental data of Fig. 2.

$F_0/10^{-5}$ N	E_0/V·mm^{-1}	R/mm	gap/mm	$\tau_R/10^{-4}$ s	$\tau_I/10^{-4}$ s
76.0	100	19.314	0.254	10.159	7.1×10^{-6}
53.5	100	19.441	0.381	3.863	3.9×10^{-7}
37.1	100	19.568	0.508	2.266	1.7×10^{-4}
42.0	80	19.314	0.254	4.405	2×10^{-10}
33.0	80	19.441	0.381	4.342	7.3×10^{-4}
22.0	80	19.568	0.508	2.646	2.1×10^{-6}

Table 2. Relaxation times, τ_R and τ_I, for obtaining good fitting with experimental data of Fig. 3.

$F_0/10^{-5}$ N	E_0/V·mm^{-1}	R/mm	gap/mm	$\tau_R/10^{-4}$ s	$\tau_I/10^{-4}$ s
4.3	100	19.314	0.254	6.171	2×10^{-10}
3.9	100	19.441	0.381	4.855	1.7×10^{-3}
2.4	80	19.441	0.381	4.352	3.6×10^{-7}
2.2	80	19.568	0.508	3.710	2×10^{-10}

Here some comments are in order. Our method introduced the conductivity ($\bar{\sigma}_\alpha$) to metallic particles. This can be understood from the charge conservation law

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \boldsymbol{j} = 0. \tag{29}$$

Here, the current can be written as Ohm's law $\boldsymbol{j} = \bar{\sigma}_\alpha \boldsymbol{E}$, while, according to the Maxwell equations, free charge $\rho = \nabla \cdot \boldsymbol{D} = \nabla \cdot \bar{\varepsilon}_\alpha \boldsymbol{E}$. Such relations naturally give birth to Eq. (6), which contains both $\bar{\sigma}_\alpha$ and $\bar{\varepsilon}_\alpha$.

To sum up, based on a first-principles approach, we have established an AC relaxation theory for dynamic metallic particles. An imaginary part has been introduced to the method established in Ref. [38] because the particle is conducting. The relaxation times were obtained according to the fitting with the experimental data. Good agreement between theory and experiment has shown that the relaxation processes of surface-polarized and free charges yield the reduction in the attracting interparticle forces between a rotating metallic particle and a stationary metallic or dielectric one. Our theory can help to determine the relaxation time of dynamic metallic particles in many fields.

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References

[1] Tan P, Tian W J, Wu J Y, Zhou L W and Huang J P 2009 *J. Phys. Chem. B* **113** 9092
[2] Zhang L F and Huang J P 2010 *Chin. Phys. B* **19** 024213
[3] Plachy T, Mrlik M, Kozakova Z, Suly P, Sedlacik M, Pavlinek V and Kuritka I 2015 *ACS Appl. Mat. Interfaces* **7** 3725
[4] Bai X K, Pu S L, Wang L W, Wang X, Yu G J and Ji H Z 2011 *Chin. Phys. B* **20** 107501
[5] He J N, Ding P, Wang J Q, Fan C Z and Liang E J 2015 *Opt. Express* **23** 6083
[6] Felicia L J and Philip J 2015 *Langmuir* **31** 3343
[7] Rabbani Y, Ashtiani M and Hashemabadi S H 2015 *Soft Matter* **31** 4453
[8] Fan C Z, Wang J Q, Zhu S M, He J N, Ding P and Liang E J 2013 *J. Opt.* **15** 055103
[9] Han Q B, Yin C, Li J, Tang Y B, Shan M L and Cao Z Q 2013 *Chin. Phys. B* **22** 094215
[10] Mestrom L, Lenders J J M, de Groot R, Hooghoudt T, Sommerdijk N A J M and Artigas M V 2015 *Nanotechnology* **26** 285602
[11] Kumar D T, Zhou Y L, Brown V, Lu X Y, Kale A, Yu L D and Xuan X C 2015 *Microfluid Nanofluid* **19** 43
[12] Vyas N S, Lawrence K L, Griffith J L and Hamzavi I H 2015 *J. Cutan. Med. Surg.* **19** 488
[13] Tao R and Sun J M 1991 *Phys. Rev. Lett.* **67** 398
[14] Halsey T C 1992 *Science* **258** 761
[15] von Pfeil K, Graham M D, Klingenberg D J and Morris J F 2002 *Phys. Rev. Lett.* **88** 188301
[16] Wen W, Huang X, Yang S, Lu K and Sheng P 2003 *Nat. Mater.* **2** 727
[17] Tzima E, Irani-Tehrani M, Kiosses W B, Dejana E, Schultz D A, Engelhardt B, Cao G Y, DeLisser H and Schwartz M A 2005 *Nature* **437** 426
[18] Misbah C 2006 *Phys. Rev. Lett.* **96** 028104
[19] Martin S, Reichert M, Stark H and Gisler T 2006 *Phys. Rev. Lett.* **97** 248301
[20] Xiao J J, Huang J P and Yu K W 2008 *J. Phys. Chem. B* **112** 6767
[21] Wang L, Wang X Q, Wang X G and Liu Y 2014 *Chin. Phys. B* **23** 025203
[22] Liu H, Tong H and Xu N 2014 *Chin. Phys. B* **23** 116105
[23] Duits M H G, Ghosh S and Mugele F 2015 *Langmuir* **31** 5689
[24] Fuhr G, Zimmermann U and Shirley S 1996 *Electromanipulation of Cells* **pp.** 259-328
[25] Becker F F, Wang X B, Huang Y, Pethig R, Vykoukal J and Gascoyne P R C 1995 *Proc. Natl. Acad. Sci. USA* **92** 860
[26] Edwards B, Mayer T S and Bhiladvala R B 2006 *Nano Lett.* **6** 626
[27] Yan Z J and Scherer N F 2013 *J. Phys. Chem. Lett.* **4** 2937
[28] Chen K, Xiang N, Quan Y L, Zhu X L, Sun D K, Yi H and Ni Z H 2014 *Microfluid Nanofluid* **16** 237
[29] He J N, Wang J Q, Ding P, Fan C Z and Liang E J 2015 *J. Opt.* **17** 055002
[30] Cebers A 2004 *Phys. Rev. Lett.* **92** 034501
[31] Dávalos-Orozco L A and del Castillo L F 1996 *J. Colloid Interface Sci.* **178** 69
[32] Fan G L, Jiang Y S, Liu L and Li F 2010 *Acta Phys. Sin.* **59** 5374 (in Chinese)
[33] Teng L H, Wang X and Lai T S 2011 *Acta Phys. Sin.* **60** 047201 (in Chinese)
[34] Lin W Z, Shou Q, Liu L N, Wu Y, Wen J H and Lai T S 2005 *Chin. Phys. Lett* **22** 188
[35] Tian W J, Liu M K and Huang J P 2007 *Phys. Rev. E* **75** 021401
[36] Tao R and Lan Y C 2005 *Phys. Rev. E* **72** 041508
[37] Tan P, Tian W J, Zhou L W and Huang J P 2009 *J. Phys. Chem. B* **113** 5412
[38] Gu G Q, Yu K W and Hui P M 2002 *J. Chem. Phys.* **116** 10989
[39] Jackson J D 1999 *Classical Electrodynamics*, 3rd edn. (New York: Wiley)
[40] Gu G Q and Yu K W 2005 *China Particuology* **3** 47
[41] Wen W, Ma H, Tam W Y and Sheng P 1997 *Phys. Rev. E* **55** R1294
[42] Russel W B, Saville D A and Schowalter W R 1989 *Colloidal Dispersions* (Cambridge, UK: Cambridge University Press)
[43] Ashby N 1975 *Am. J. Phys.* **43** 553