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Time-Varying Plasmonic Particles

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***Abstract* – In this paper, we derive a third-order linear differential equation for describing polarization dynamics of a spherical plasmonic nanoparticle. We entitle such equation, being analogous to the Abraham-Lorentz equation, as the Rüdenberg equation that was initially written in 1907 for elucidating the Hertzian dipole antenna in the receiving regime. Based on this equation, we study the case when the plasma frequency of the nanoparticle material varies in time. Under some assumptions, we find analytical solutions which describe the radiative decay of such time-varying nanoparticle.**

I. INTRODUCTION

Temporal modulation is an additional degree of freedom for engineering electromagnetic systems [1] which opens up novel possibilities, results in exceeding conventional limitations [2], and allows us to design efficient systems with various functionalities such as nonreciprocity [3]. Recently, we have made some steps towards fundamental understanding of basics and principles underlying this re-emerging research direction. In particular, we initially attempted to describe a meta-atom which is modulated in time [4], and, subsequently, scrutinized the instantaneous radiation effects [5] and the polarizability of such time-varying meta-atoms [6]. As a continuation of these efforts, in this paper, we contemplate a time-varying spherical plasmonic nanoparticle located in free space and investigate how time modulation of the plasma frequency of its plasmonic material will affect the interaction of electromagnetic waves with the nanoparticle. Particularly, we focus on studying the (radiative) decay of an excited nanoparticle which is under such temporal modulation. We hope that this work will help to better understand the response of nonstationary (time-varying) plasmonic nanoparticles and other time-modulated resonant scatterers.

II. RÜDENBERG EQUATION

Let us start from writing the quasi-static polarizability (α_{qs}) of a stationary plasmonic nanoparticle in the frequency domain. For a spherical nanoparticle, it is known that $\alpha_{\mathrm{qs}} = 3\epsilon_0 V(\epsilon_{\mathrm{r}}-1)/(\epsilon_{\mathrm{r}}+2)$, in which ϵ_0 is the free-space permittivity, V refers to the nanoparticle volume, and ϵ_{r} represents the relative permittivity of the nanoparticle material. We model this effective macroscopic parameter, ϵ_{r}, by the Drude model which assumes no restoring spring force for the electrons: $\epsilon_{\mathrm{r}} = 1-\omega_{\mathrm{p}}^2/(\omega^2-j\omega\Gamma)$. Here, ω_{p} and Γ denote the plasma frequency and the collision frequency, respectively. By substituting the above expression for the relative permittivity in the first formula describing the quasi-static polarizability, we deduce that $1/\alpha_{\mathrm{qs}} = (-\omega^2+j\omega\Gamma+\omega_{\mathrm{p}}^2/3)/(\epsilon_0 V\omega_{\mathrm{p}}^2)$. Based on the definition, the induced electric dipole moment $\mathbf{p}$ is linked to the excitation electric field $\mathbf{E}$ as $(1/\alpha_{\mathrm{qs}})\mathbf{p} = \mathbf{E}$. Consequently, we readily write that $(\epsilon_0 V\omega_{\mathrm{p}}^2)^{-1}(-\omega^2\mathbf{p}+j\omega\Gamma\mathbf{p}+(\omega_{\mathrm{p}}^2/3)\mathbf{p}) = \mathbf{E}$. Next, we make the inverse Fourier transform in order to achieve a linear differential equation which describes the induced dipole moment. The result reads

$$(\epsilon_0 V\omega_{\mathrm{p}}^2)^{-1}\left[\frac{d^2\mathbf{p}(t)}{dt^2}+\Gamma\frac{d\mathbf{p}(t)}{dt}+\frac{\omega_{\mathrm{p}}^2}{3}\mathbf{p}(t)\right] = \mathbf{E}(t). \tag{1}$$

However, this equation is deficient in one fundamental respect, and that is the "radiation reaction". We need to add the corresponding electric field radiated by the dipole (self-action) that defines the radiation reaction. This component, explicitly derived in Ref. [5], is proportional to the third derivative of the dipole moment and is

given by $\mathbf{E}_{\rm rad-reac} = [\mu_0/(6\pi c)]d^3\mathbf{p}/dt^3$. Therefore, by taking this component into account, and after some simplifications, Eq. (1) is modified as

$$-\frac{\mu_0}{6\pi c}(\epsilon_0 V\omega_{\rm p}^2)\frac{d^3\mathbf{p}(t)}{dt^3}+\frac{d^2\mathbf{p}(t)}{dt^2}+\Gamma\frac{d\mathbf{p}(t)}{dt}+\frac{\omega_{\rm p}^2}{3}\mathbf{p}(t)=(\epsilon_0 V\omega_{\rm p}^2)\mathbf{E}(t). \tag{2}$$

Notice that, very importantly, if we now transform this equation back to the frequency domain, replacing d/dt by $j\omega$, we find the inverse polarizability in the famous form $1/\alpha = (1/\alpha_{\rm qs}) + jk_0^3/(6\pi\epsilon_0)$, where k_0 is the free-space wavenumber. This means that the scattering loss term in this form is valid not only for time-harmonic fields but properly models instantaneous radiated power for arbitrary time variations of the dipole moment.

As expected, Eq. (2) is similar to the Abraham–Lorentz equation that also includes the radiation reaction term. Indeed, comparing with that equation, $-q^2[\mu_0/(6\pi cm)]d^3\mathbf{r}/dt^3 + d^2\mathbf{r}/dt^2 + \omega_0^2\mathbf{r} = 0$, conspicuously, one sees a clear analogy (note that $\mathbf{r}$ is the displacement, q is the electron charge, and m represents the electron mass). Furthermore, importantly, what we derived in Eq. (2) is absolutely in agreement with what R. Rüdenberg wrote in 1907 for a receiving Hertzian dipole (only about 18 years after the paper by H. Hertz written in 1889 describing the dipole). Based on equation (8) in Ref. [7], Rüdenberg finds: $-Sd^3e/dt^3 + Ld^2e/dt^2 + Rde/dt + (1/C)e = lE$. In this equation, e is the electric charge, L and C are the effective inductance and capacitance measuring the magnetic and electric energies stored around the dipole, and R is the resistance for taking the ohmic losses into account. The parameter S, associated with the radiation reaction term, determines what Rüdenberg calls "Strahlungswiderstand" or radiation resistance. Since the dipole moment is the multiplication of the electric charge and the dipole length, therefore, we modify the initial version of Rüdenberg's equation and write

$$-\frac{\mu_0}{6\pi c}\frac{l^2}{L}\frac{d^3\mathbf{p}}{dt^3}+\frac{d^2\mathbf{p}}{dt^2}+\frac{R}{L}\frac{d\mathbf{p}}{dt}+\frac{1}{LC}\mathbf{p}=\frac{l^2}{L}\mathbf{E}. \tag{3}$$

This equation is precisely the same as Eq. 2 that we derived for the plasmonic nanoparticle. Indeed, in Eq. (3), we only need to replace l^2/L by $\epsilon_0 V\omega_{\rm p}^2$ ($l^2/L \to \epsilon_0 V\omega_{\rm p}^2$), $R/L \to \Gamma$, and, finally, $1/(LC) \to \omega_{\rm p}^2/3$. Thus, let us entitle Eq. (2) as Rüdenberg equation (RE).

Next, we can assume that the coefficients and parameters in the RE are time dependent. The equation will remain a linear differential equation allowing simple analytical solutions. By using this assumption, in the next section, we will show these solutions which describe the decay of an excited plasmonic nanoparticle that is modulated externally in time. For that, we will suppose that only the plasma frequency varies in time (i.e., $\omega_{\rm p} = \omega_{\rm p}(t)$), while the volume of the nanoparticle and the collision frequency are constant. Note that the volume can be also considered mutable in time. However, in practice, it may be easier to change the plasma frequency.

III. Studying decay process

In order to investigate the decay process and see how it is affected by temporally changing the plasma frequency, we choose the excitation field equal to zero (resulting in a homogeneous equation) and assume that the nanoparticle is excited at zero moment of time by a delta pulse. However, because the order of RE is three, it is not straightforward to analytically solve the equation and study its salient properties. Therefore, we need to do some assumptions and attempt to reduce the order to at least two. To do that, we suppose that the radiation reaction term, associated with the third time derivative, and the dissipative term, related to the first time derivative, are much smaller than the last term in the equation. In other words, $-(\mu_0/6\pi c)(\epsilon_0 V\omega_{\rm p}^2)d^3\mathbf{p}/dt^3 \ll (\omega_{\rm p}^2/3)\mathbf{p}$ and $\Gamma d\mathbf{p}/dt \ll (\omega_{\rm p}^2/3)\mathbf{p}$. Accordingly, neglecting those terms, we conclude that $d^2\mathbf{p}/dt^2 + (\omega_{\rm p}^2/3)\mathbf{p} = 0$, which results in the following expression: $d^3\mathbf{p}/dt^3 = -(1/3)\big[2\omega_{\rm p}(d\omega_{\rm p}/dt)\mathbf{p} + \omega_{\rm p}^2 d\mathbf{p}/dt\big]$. This approximation is indeed helpful. Now, it is only sufficient to supersede this into the main Rüdenberg equation and rewrite it. This time, the order of our linear differential equation is two, as desired. After some algebraic manipulations, we finally deduce that

$$\frac{d^2\mathbf{p}}{dt^2}+\left[\Gamma+\frac{\mu_0}{6\pi c}(\epsilon_0 V\omega_{\rm p}^2)\frac{\omega_{\rm p}^2}{3}\right]\frac{d\mathbf{p}}{dt}+\frac{\omega_{\rm p}^2}{3}\left[1+\frac{\mu_0}{6\pi c}(\epsilon_0 V\omega_{\rm p}^2)\frac{2}{\omega_{\rm p}}\frac{d\omega_{\rm p}}{dt}\right]\mathbf{p}=0. \tag{4}$$

Let us consider the following identity:

$$\mathbf{p}(t)=\exp\left[-\frac{1}{2}\int\Big(\Gamma+\frac{\mu_0}{6\pi c}(\epsilon_0 V\omega_{\rm p}^2)\frac{\omega_{\rm p}^2}{3}\Big)dt\right]\mathbf{p}_{\rm a}(t), \tag{5}$$

and substitute it into Eq. (4). This identity, which introduces the auxiliary vector $\mathbf{p}_{\rm a}(t)$, is used in the theory of differential equations to remove the first time derivative. Indeed, after substitution, we find a second-order differential equation without the first time derivative:

$$\frac{d^2\mathbf{p}_{\rm a}}{dt^2}+\left[\frac{1}{3}\omega_{\rm p}^2-\frac{1}{6}\left(\frac{V}{6\pi c^3}\right)\omega_{\rm p}^4\Gamma-\frac{1}{36}\left(\frac{V}{6\pi c^3}\right)^2\omega_{\rm p}^8-\frac{1}{4}\Gamma^2\right]\mathbf{p}_{\rm a}=0. \tag{6}$$

At this point, we need to determine how the plasma frequency should vary in time. One possibility is to assume that $\omega_{\rm p}$ changes periodically. Let us choose the conventional harmonic variation so that $\omega_{\rm p}(t)=\Omega_{\rm p}\cdot f(t)$ where $f(t)=\sqrt{1+\delta\cos(\omega_{\rm m}t+\phi_{\rm m})}$. The reason behind using the square root is obvious from the above equation in which we have only even powers of the plasma frequency. With this specific temporal profile for $\omega_{\rm p}$, if δ, which denotes the modulation strength, is small compared to unity, we can write the following approximations: $\omega_{\rm p}^4(t)\approx\Omega_{\rm p}^4(1+2\delta\cos(\omega_{\rm m}t+\phi_{\rm m}))$ and $\omega_{\rm p}^8(t)\approx\Omega_{\rm p}^8(1+4\delta\cos(\omega_{\rm m}t+\phi_{\rm m}))$. By employing these two useful approximations, Eq. (6) reduces to

$$\frac{d^2\mathbf{p}_{\rm a}}{dt^2}+\Big(a-2b\cos(\omega_{\rm m}t+\phi_{\rm m})\Big)\mathbf{p}_{\rm a}=0, \tag{7}$$

in which

$$a=\frac{1}{3}\Omega_{\rm p}^2-\frac{1}{6}\left(\frac{V}{6\pi c^3}\right)\Gamma\Omega_{\rm p}^4-\frac{1}{36}\left(\frac{V}{6\pi c^3}\right)^2\Omega_{\rm p}^8-\frac{1}{4}\Gamma^2,\quad b=\frac{\delta}{18}\left(\frac{V}{6\pi c^3}\right)^2\Omega_{\rm p}^8+\frac{\delta}{6}\left(\frac{V}{6\pi c^3}\right)\Gamma\Omega_{\rm p}^4-\frac{\delta}{6}\Omega_{\rm p}^2.$$

As a sanity check, if $\delta=0$, we see that $b=0$, and, therefore, $\mathbf{p}_{\rm a}$ is oscillating with an angular frequency $\omega_0=\sqrt{a}$. Note that in this case, as expected, ω_0 is different from $\Omega_{\rm p}/\sqrt{3}$. Clearly, this difference stems from the additional terms that are contributing into the parameter a and are in fact associated with a nonzero collision rate and with the radiation damping. Also, notice that while $\mathbf{p}_{\rm a}$ is oscillating, the dipole moment $\mathbf{p}$ decays in time due to the presence of the exponential function which is multiplied by $\mathbf{p}_{\rm a}$ (see Eq. (5)). Thus, the decay rate of the induced dipole in this case ($\delta=0$) is related only to this exponential factor.

As the last step, Eq. (7) can be modified by changing the variable as $2u=\omega_{\rm m}t+\phi_{\rm m}$. If we impose this change, the modified version of Eq. (7) reads $d^2\mathbf{p}_{\rm a}/du^2+\big([a/(\omega_{\rm m}^2/4)]-2[b/(\omega_{\rm m}^2/4)]\cos(2u)\big)\mathbf{p}_{\rm a}=0$. This differential equation is Mathieu's differential equation whose solutions are known as Mathieu functions.

IV. Conclusion

We initially derived the Rüdenberg differential equation for a spherical plasmonic nanoparticle, and then assumed that the plasma angular frequency varies in time. By imposing the condition that the radiation reaction and the dissipative terms are small enough (compared to the natural frequency term), we showed that it is possible to analytically find the radiative decay of an excited state of the modulated nanoparticle in terms of an exponential factor which is multiplied by an important auxiliary vector described by Mathieu functions.

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