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# Giant dipole moment induced by an interface

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## ABSTRACT

When a small particle is illuminated by a laser beam, an oscillating electric dipole moment is induced. When located near an interface, the reflected laser light will change the local electric field, and thereby the dipole moment. However, also the dipole radiation, emitted by the particle, reflects at the interface, and contributes to the electric field at the location of the particle. This reflected field alters the dipole moment. We find that, under certain circumstances, the induced dipole moment by the dipole's own radiation far exceeds the dipole moment induced by the laser.

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An oscillating electric dipole is the most extensively studied source of electromagnetic radiation, both theoretically and experimentally, and both classically and quantum mechanically. Any radiating object with dimensions smaller than a wavelength is in good approximation an electric dipole. This includes atoms, molecules and nanoparticles when it comes to visible light, but for larger wavelengths also macroscopic radiators can approximately be electric dipoles. In his landmark publication in 1909 [1], Sommerfeld considered the propagation of radio waves, emitted by a small antenna, over the surface of the Earth. In a more recent application, low frequency dipole radiation was used to measure to profile of the ocean floor [2]. Interestingly, the surface not only reflects the radiation, but the reflected light can alter the emission properties of the dipole. Both the radiation pattern (angular distribution) and the overall emitted power are influenced by the presence of the surface. The pioneering work by Chance and coworkers [3] predicted that the emitted power by a dipole depends not only on the state of oscillation of the dipole, but also on its distance to the surface. In a series of experiments by Drexhage [4], this distance dependence of the power emission was confirmed for monomolecular layers on a substrate.

We shall consider the setup shown in Fig. 1, where a small spherical dielectric particle is irradiated by a laser beam with angular frequency  $\omega$ . The induced dipole moment is then

$$\mathbf{d}(t) = \operatorname{Re}\left[\mathbf{d}\exp(-i\omega t)\right],\tag{1}$$

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Fig. 1. Shown is the setup under consideration. A small dielectric particle is located near an interface, and irradiated by a laser beam.

with **d** the complex amplitude. The particle has radius *R*, (relative) permittivity  $\varepsilon_p$ , and it has a distance *H* to the surface of a substrate. The particle is embedded in a medium with permittivity  $\varepsilon_1$  and permeability  $\mu_1$ , both assumed to be positive. The index of refraction of the embedding material is  $n_1 = \sqrt{\varepsilon_1 \mu_1}$ . The material of the substrate has permittivity  $\varepsilon_2$  and permeability  $\mu_2$ , and we shall put no restrictions on these parameters.

The time-averaged power, emitted as dipole radiation by the particle, located at  $\mathbf{r}_{o}$ , is given by [5]

$$P_{\rm e} = \frac{1}{2}\omega \,{\rm Im}\,\mathbf{d}^* \cdot \big[\mathbf{E}_{\rm s}(\mathbf{r}_{\rm o}) + \mathbf{E}_{\rm r}(\mathbf{r}_{\rm o})\big]. \tag{2}$$

Here,  $\mathbf{E}_{s}(\mathbf{r})$  is the complex amplitude of the electric field of the radiation emitted by the dipole itself (source field) and  $\mathbf{E}_{r}(\mathbf{r})$  is the





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field reflected off the interface. The source field gives a contribution of

$$P_{\rm s} = \mu_1 n_1 \omega \frac{k_0^3}{12\pi\varepsilon_0} \mathbf{d}^* \cdot \mathbf{d}.$$
 (3)

Here,  $k_0 = \omega/c$  is the wave number in free space. The reflected electric field  $\mathbf{E}_{\mathbf{r}}(\mathbf{r})$  adds to the source field in Eq. (2), and this leads to a modification of the power emission. Clearly, this reflected field, evaluated at  $\mathbf{r}_0$ , depends on the distance to the surface, and thus leads to a distance dependent power emission. This celebrated phenomenon has been studied by many authors, both experimentally [6–13] and theoretically [14–22].

The laser beam reflects at the interface, and the reflected radiation interferes with the original beam. We shall indicate by  $\mathbf{E}_{L+R}(\mathbf{r})$  the sum of the two. For a spherical particle with dimensions much smaller than a wavelength ( $k_0 R \ll 1$ ), the dipole moment **d** is induced by the external field as

$$\mathbf{d} = \alpha \left[ \mathbf{E}_{L+R}(\mathbf{r}_{o}) + \mathbf{E}_{r}(\mathbf{r}_{o}) \right], \tag{4}$$

with  $\alpha$  the polarizability of the particle. It is common to assume that only  $\mathbf{E}_{L+R}(\mathbf{r}_o)$  determines the dipole moment, so that **d** is a given for a problem, and is not influenced by the presence of the surface (other than through the reflected laser light). The addition of  $\mathbf{E}_r(\mathbf{r}_o)$  to the induced dipole moment implies that **d** is modified by its own reflected field, and since this field at  $\mathbf{r}_o$  depends on the location of the dipole, the induced **d** also depends on the distance between the particle and the surface. We shall show below that the contribution of  $\mathbf{E}_r(\mathbf{r}_o)$  to **d** is indeed minimal for most practical purposes, but that under certain circumstances the effect of the reflected dipole radiation on the induced **d** can be enormous.

It seems that we can just add  $\mathbf{E}_{r}(\mathbf{r}_{o})$  to  $\mathbf{E}_{L+R}(\mathbf{r}_{o})$  in Eq. (4), and then see which contribution dominates. The situation is, however, more intricate. The reflected dipole field  $\mathbf{E}_{r}(\mathbf{r})$  is generated by the oscillating dipole. This field then affects the dipole moment **d** which was responsible for the emission in the first place. So, the reflected field alters its own source. In order to determine the contribution of  $\mathbf{E}_{r}(\mathbf{r})$  to the induced **d**, we first compute the reflected field in  $\mathbf{r}_{o}$ . We adopt an angular spectrum representation of the emitted dipole radiation [23]. This is a superposition of plane waves. The reflection of each plane wave can be accounted for by a Fresnel reflection coefficient, and the reflected dipole field then becomes a superposition of reflected plane waves. When evaluated at  $\mathbf{r}_{o}$ , we obtain the representation

$$\mathbf{E}_{\mathrm{r}}(\mathbf{r}_{\mathrm{o}}) = \frac{i\mu_{1}k_{\mathrm{o}}}{8\pi^{2}\varepsilon_{\mathrm{o}}} \sum_{\sigma} \int \mathrm{d}^{2}\mathbf{k}_{||} \frac{1}{\nu_{1}} e^{2i\nu_{1}h} (\mathbf{d} \cdot \mathbf{e}_{\sigma}) R_{\sigma} \mathbf{e}_{\sigma,\mathrm{r}}.$$
 (5)

Here, the summation is over the polarization types  $\sigma = s$ , p (surface and plane polarized), and the integral runs over the  $\mathbf{k}_{||}$  plane, which is the *xy* plane. This vector  $\mathbf{k}_{||}$  is the parallel component of an incident wave vector, and we introduce  $u = k_{||}/k_0$  for its normalized magnitude. The variable  $v_1$  comes from the *z* component of an incident wave, and is given by  $v_1 = \sqrt{n_1^2 - u^2}$ . For  $0 \le u < n_1$  the incident wave is traveling, and for  $u > n_1$  the incident wave is evanescent. The unit vectors  $\mathbf{e}_s$  and  $\mathbf{e}_p$  are the polarization vectors for incident *s* and *p* polarized waves, and similarly,  $\mathbf{e}_{s,r}$  and  $\mathbf{e}_{p,r}$  are the polarization vectors for reflected waves. The Fresnel reflection coefficients  $R_{\sigma}$  are functions of *u*. We have introduced  $h = k_0 H$  for the dimensionless distance between the particle and the surface. On this scale, a distance of  $2\pi$  corresponds to a free-space optical wavelength.

At this point we shall assume that the laser is *s* polarized (electric field parallel to the surface). This is not a necessary restriction, but it simplifies the formulas considerably for the present purpose.



**Fig. 2.** The figure shows the real (solid curve) and imaginary (dashed curve) parts of the function v(h) for  $\varepsilon_1 = 1$  and  $\varepsilon_2 = 6$ .

Then **d** is parallel to the surface as well. The angular integration in Eq. (5) can be performed in closed form. This yields for the reflected field at  $\mathbf{r}_{0}$ :

$$\mathbf{E}_{\mathrm{r}}(\mathbf{r}_{\mathrm{o}}) = \frac{i(n_{1}k_{\mathrm{o}})^{3}}{8\pi\varepsilon_{0}\varepsilon_{1}}v(h)\mathbf{d}.$$
(6)

The function v(h) is found to be

$$v(h) = \frac{1}{n_1^3} \int_0^\infty \mathrm{d} u \frac{u}{v_1} e^{2iv_1 h} [n_1^2 R_s(u) - v_1^2 R_p(u)].$$
(7)

A typical example of v(h) is shown in Fig. 2. The real and imaginary parts diverge in general for  $h \rightarrow 0$ . For larger h, the real and imaginary parts become oscillatory. We shall set  $\mu_1 = \mu_2 = 1$  for all figures, since the permeabilities have as good as no effect.

The laser is assumed to be s polarized, and we shall take the x axis along the polarization direction. Then the reflected field is also s polarized, and we can write

$$\mathbf{E}_{\mathrm{L+R}}(\mathbf{r}_{\mathrm{o}}) = E_{\mathrm{o}}\mathbf{m}(h),\tag{8}$$

where  $E_0$  is the amplitude, and the mode-structure function  $\mathbf{m}(h)$  at the location of the dipole is

$$\mathbf{m}(h) = \left(e^{-ihn_1\cos\theta_i} + R_s e^{ihn_1\cos\theta_i}\right)\mathbf{e}_x,\tag{9}$$

with  $\theta_i$  the angle of incidence. For the particle, we introduce the dimensionless polarizability volume

$$\bar{\mathcal{V}}_{\rm p} = \frac{(n_1 k_0)^3}{4\pi \varepsilon_0 \varepsilon_1} \alpha,\tag{10}$$

in terms of which we have

$$\alpha \mathbf{E}_{\mathbf{r}}(\mathbf{r}_{0}) = \frac{i}{2} \bar{\mathcal{V}}_{\mathbf{p}} \mathbf{v}(h) \mathbf{d}.$$
(11)

We now substitute  $\mathbf{E}_{L+R}(\mathbf{r}_0) + \mathbf{E}_r(\mathbf{r}_0)$  in Eq. (4). We thus obtain

$$\mathbf{d} = \alpha E_{\mathbf{o}} \mathbf{m}(h) + \frac{i}{2} \mathbf{v}(h) \bar{\mathcal{V}}_{\mathbf{p}} \mathbf{d}.$$
 (12)

This is an equation for the dipole moment  $\mathbf{d}$ , with solution

$$\mathbf{d} = \Upsilon(h)\mathbf{m}(h)\alpha E_0. \tag{13}$$

Here we have introduced the resolvent function

$$\Upsilon(h) = \frac{1}{1 - (i/2)\nu(h)\bar{\mathcal{V}}_{p}}.$$
(14)

The contribution of the reflected dipole radiation to **d** is incorporated in the function  $\Upsilon(h)$ . Any deviation of this function from unity is due to the reflected dipole waves. We see from Eq. (13) that  $\mathbf{E}_{r}(\mathbf{r}_{o})$  does not just add to the laser contribution  $\mathbf{E}_{L+R}(\mathbf{r}_{o})$ . The polarizability of the particle comes in through  $\bar{\mathcal{V}}_{p}$ , which appears



**Fig. 3.** The graph shows the real (solid curve) and imaginary (dashed curve) parts of the function  $\Upsilon(h)$  for  $\varepsilon_1 = 1$ ,  $\varepsilon_2 = 6$ ,  $\varepsilon_p = 3$  and  $\bar{R} = 2.5$  The vertical dashed line is at  $h = \bar{R}$ .

in the denominator in Eq. (14). The dipole moment **d** is still proportional to the laser amplitude  $E_0$ . For h large, we have  $v(h) \rightarrow 0$ , as can be seen from Fig. 2, and so  $\Upsilon(h) \rightarrow 1$  when the dipole is far away from the interface.

In the derivation of the result (13) we have not made any assumptions about the particle (other than that its polarizability is a scalar, rather than a tensor). The scattering of a plane wave by a spherical particle has an exact solution, known as Mie theory. It follows from this exact solution that in the limit where the radius of the particle is much less than a wavelength, the particle is in good approximation an electric dipole [24]. Its polarizability is  $\eta 4\pi \varepsilon_0 \varepsilon_1 R^3$ , and with Eq. (10) we find for the dimensionless polarizability volume

$$\bar{\mathcal{V}}_{\mathrm{p}} = \eta (n_1 \bar{R})^3. \tag{15}$$

Here,  $\bar{R} = k_0 R$  is the dimensionless radius of the particle, and

$$\eta = \frac{\varepsilon_{\rm p} - \varepsilon_{\rm 1}}{\varepsilon_{\rm p} + 2\varepsilon_{\rm 1}}.\tag{16}$$

The advantage of using  $\bar{\mathcal{V}}_p$  here, rather than  $\alpha$ , can be seen from Eq. (15). Both  $\eta$  and  $\bar{R}$  are of the order of unity, and therefore both  $\bar{\mathcal{V}}_p$  and  $\Upsilon(h)$  are of the order of unity, and they are dimensionless. For instance, for R = 100 nm and  $\lambda = 500$  nm we have  $\bar{\mathcal{V}}_p \sim 2$ . The dielectric constant  $\varepsilon_p$  of the particle can be complex (absorption), which would make  $\eta$  complex. For  $\varepsilon_p$  real and  $\varepsilon_1 > \varepsilon_p$  we have  $\eta < 0$ , and therefore  $\bar{\mathcal{V}}_p < 0$ .

In order to reveal the significance of the contribution of  $\Upsilon(h)$ to the induced dipole moment, we consider some numerical examples. The resolvent  $\Upsilon(h)$  is defined for h > 0, but we see from Fig. 1 that the range should be restricted to  $h > \overline{R}$  (the particle would touch the surface for  $h = \overline{R}$ ). Fig. 3 shows a typical graph of the real and imaginary parts of  $\Upsilon(h)$ , for the parameters given in the caption. Without the reflected dipole radiation contribution to **d**, we would have Re  $\Upsilon(h) = 1$  and Im  $\Upsilon(h) = 0$ . We find an oscillatory deviation from  $\Upsilon(h) = 1$ , but the deviations are marginal. The value of  $\overline{R}$  is taken as 2.5. This corresponds to a particle radius of 0.4 times an optical wavelength. It can be shown that for smaller values of  $\overline{R}$  any effect disappears. For larger values, the long wavelength limit becomes questionable. We shall take this borderline value of  $\bar{R} = 2.5$  in all further examples. Fig. 4 show  $|\Upsilon(h)|$  for  $\varepsilon_1 = 5$ ,  $\varepsilon_2 = 1$  and  $\varepsilon_p = 2$ . The embedding medium is denser than the particle, and we have  $\eta = -0.25$ ,  $\bar{\mathcal{V}}_p = -43.7$ . A large peak appears at  $h \approx 3.5$ , and smaller peaks appear at larger h values. The peak height is about  $|\Upsilon(h)| \approx 10$ , so the induced dipole moment has increased by a factor of 10 due to the contribution of the reflected dipole radiation.

It appears that the resolvent  $\Upsilon(h)$  displays peaks, which can be moderate, as in Fig. 3, or very pronounced, as in Fig. 4. In order to elucidate the source of this phenomenon we consider the case where the substrate is a perfect conductor. For such a material,



**Fig. 4.** The figure shows  $|\Upsilon(h)|$  for  $\varepsilon_1 = 5$ ,  $\varepsilon_2 = 1$  and  $\varepsilon_p = 2$ , as a function of *h*, and we see that large peaks appear in the resolvent function.



**Fig. 5.** The figure shows  $|\Upsilon(h)|$  for a perfect conductor,  $\varepsilon_1 = 1$  and  $\varepsilon_p = 3$ , as a function of *h*.

the Fresnel reflection coefficients are  $R_s = -1$  and  $R_p = 1$ , and the integral in Eq. (7) can be evaluated in closed form. We find

$$v(h) = \frac{2i}{\beta} \left( 1 + \frac{i}{\beta} - \frac{1}{\beta^2} \right) e^{i\beta},\tag{17}$$

with  $\beta = 2n_1h$ . Fig. 5 shows a typical graph for a perfect conductor. For not too small distances *h*, we only need to retain the term  $(2i/\beta) \exp(i\beta)$ . Then the resolvent becomes

$$\Upsilon(h) \approx \frac{\beta}{\beta + \bar{\mathcal{V}}_{p} \exp(i\beta)}.$$
(18)

The oscillatory behavior comes from  $\exp(i\beta)$  in the denominator. This function is periodic with  $\Delta\beta = 2\pi$ , so the peaks are spaced with  $\Delta h \approx \pi/n_1$ . For  $\varepsilon_1 = 1$  and  $\varepsilon_p = 3$ , as in Fig. 5, we have  $n_1 = 1$ ,  $\eta = 0.4$  and  $\bar{\mathcal{V}}_p = 6.25$ . Since  $\bar{\mathcal{V}}_p$  is positive, the maxima appear at  $\exp(i\beta_n) \approx -1$ . This gives  $\beta_n \approx \pi (2n + 1)$  and  $h_n \approx \pi (n + 1/2)/n_1$ . For Fig. 5 we then find for the peak locations  $h_1 \approx 4.7$ ,  $h_2 \approx 7.9$  and  $h_3 \approx 11$ , which is in good agreement with the graph. The approximate peak heights follow from Eq. (18) with  $\exp(i\beta_n) = -1$ , and we get  $\Upsilon(h_1) \approx 3.0$ ,  $\Upsilon(h_2) \approx 1.7$  and  $\Upsilon(h_3) \approx 1.4$ , also in good agreement with the graph. We conclude that the appearance of the peaks in  $\Upsilon(h)$  is due to the oscillatory nature of  $\nu(h)$  for h moderate to large.

The solution for the induced dipole moment **d** is given by Eq. (13) for *s* polarization. The magnitude of this complex-valued vector is  $d_0 = \sqrt{\mathbf{d}^* \cdot \mathbf{d}}$ . Without the surface, this would be  $d_0 = |\alpha|E_0$ . The mode-structure function  $\mathbf{m}(h)$  accounts for the interference between the reflected laser light and the incident light, whereas the resolvent  $\Upsilon(h)$  represents the effect on the induced dipole moment by its own reflected field. Fig. 6 shows  $d_0/(|\alpha|E_0)$  for  $\varepsilon_1 = 5$ ,  $\varepsilon_2 = 1$ ,  $\varepsilon_p = 2$  and an angle of incidence of 30°. These are the same parameters as for  $|\Upsilon(h)|$  in Fig. 4. The dotted horizontal line at unity is what the dipole moment would be without the interface, and the dashed curve is what the dipole moment would be if only the laser and its reflection would induce the dipole moment. The solid curve represents the dipole moment when the contribution from the reflected dipole radiation is included. There is a giant spike at  $h \approx 3.5$ , and this represents an amplification of



**Fig. 6.** The figure shows  $d_0/(|\alpha|E_0)$  for  $\varepsilon_1 = 5$ ,  $\varepsilon_2 = 1$ ,  $\varepsilon_p = 2$  and  $\theta_i = 30^\circ$ , as a function of *h*.

about 15, when compared to excitation by the laser only. Smaller peaks appear at larger *h* values. Even the smaller peak at about  $h \approx 5$  still shows an enhancement of about 7. Clearly, the contribution of the reflected dipole radiation to its own source can be huge for certain discrete values of *h*. It should be noted that this phenomenon is the exception rather than a universal feature. For most combinations of parameters, the dashed and solid curves are very close to each other, although some weak peaks are always present due to the oscillations of function v(h).

The dipole moment **d**, and its *h* dependence, can be observed experimentally through the emitted power. The particles can be deposited on a spacer layer with a controlled thickness *h*, and the emitted power can be observed as a function of the layer thickness [4]. The contribution of the source field to the power is given by Eq. (3), with **d** given by the right-hand side of Eq. (13). This gives for the source contribution

$$P_{\rm s} = \mu_1 n_1 P_{\rm f}(\mathbf{m}^* \cdot \mathbf{m}) |\Upsilon|^2, \tag{19}$$

where we have set

$$P_{\rm f} = \omega \frac{k_0^3}{12\pi\varepsilon_0} |\alpha|^2 E_0^2. \tag{20}$$

This equals the power emitted by the dipole in free space, and  $\mu_1 n_1 P_f$  is the power emitted by the particle in the embedding medium, but without an interface. The contribution of  $\mathbf{E}_r(\mathbf{r}_0)$  to the emitted power follows from substituting the right-hand side of Eq. (6) into Eq. (2). Combining the two contributions then yields the final result for the emitted power:

$$P_{\mathsf{e}}(h) = \mu_1 n_1 P_{\mathsf{f}} \big[ \mathbf{m}(h)^* \cdot \mathbf{m}(h) \big] \big| \Upsilon(h) \big|^2 w(h).$$
(21)

Here, the function w(h) accounts for the contribution of the reflected field to the power emission, and is given by [25]

$$w(h) = 1 + \frac{3}{4} \operatorname{Re} v(h).$$
 (22)

Fig. 7 illustrates the emitted power for  $\varepsilon_1 = 1$  (vacuum),  $\varepsilon_3 = 0$  (epsilon-near-zero material),  $\varepsilon_p = 3$ , and a 30° angle of incidence. The dotted line at unity represents the power that would be emitted by the particle in the absence of the interface. The dashed curve shows the emitted power if only the laser and its reflection would induce the dipole moment. The solid curve shows the result if the contribution of the reflected dipole radiation to the induced dipole moment is taken into account. A sharp peak appears at  $h \approx 3$ , and the power is enhanced by about a factor of 13 due to the effect of the reflected dipole radiation on **d**.

When a small particle, located near an interface, is irradiated by a laser beam, an electric dipole moment **d** is induced. The oscillating dipole emits radiation, which is amenable to experimental observation. As for orders of magnitude, at 500 nm and with a moderate laser intensity of about  $10^4 \text{ W/m}^2$ , the emitted power in free space is  $P_{\rm f} \sim 10^{-9}$ W, which is about  $5 \times 10^9$  photons per



**Fig. 7.** Shown is the emitted power for  $\varepsilon_1 = 1$ ,  $\varepsilon_2 = 0$ ,  $\varepsilon_p = 3$  and  $\theta_i = 30^\circ$ , as a function of *h*.

second. The dipole moment is induced by the local electric field, which is under most circumstances in good approximation the electric field of the laser. However, also the dipole radiation that is reflected by the surface contributes to the local electric field. This leads to a multiplicative factor  $\Upsilon(h)$  in Eq. (13). Typical graphs of this function are shown in Figs. 3 and 4. Any deviation of  $\Upsilon(h)$ from unity is due to the reflected dipole radiation. We see from Fig. 4 that a large peak appears for a relatively small h value, and the peak height is of the order of 10. This represents an enhancement of an order of magnitude in the induced dipole moment, as compared to its value for laser irradiation only. This huge peak is due to the peculiar appearance of the polarizability  $\alpha$  of the particle in the resolvent  $\Upsilon(h)$ . It appears in the denominator in Eq. (14) through  $\bar{\mathcal{V}}_{p}$ . Since the function v(h), multiplying  $\alpha$ , is oscillatory, as seen in Fig. 2, there is a possibility for the appearance of giant peaks in the value of the dipole moment for certain distances h between the dipole and the interface. An increase in dipole moment goes together with an increase in emitted power. This power is absorbed from the laser beam. In solar cells, the power is absorbed from the sunlight. By covering the cell's surface with nanoparticles, absorption could be enhanced, and this would increase the efficiency of the energy conversion. If the particles could be tuned to one of the giant peaks that we predict, this could lead to a significant improvement of the device.

## **CRediT** authorship contribution statement

This work was performed solely by Henk F. Arnoldus.

### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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