Light absorption by a dipole

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In semiclassical radiation theory, the electric dipole moment induced on an atom by a strong incident field, absorbs much more energy, per sec, than is flowing through its geometrical cross section. This means, the atom has the capability to "suck up" electromagnetic energy from a spatial region that is by far larger than its own volume. An intuitive understanding of this effect is provided by studying, in the framework of classical electrodynamics, the energy flow in the total field made up by superposition of the incident wave and the field that is generated by the dipole also in the absorptive case.

I. INTRODUCTION

In the so-called semiclassical radiation theory the atoms are described quantum mechanically, while the radiation field is considered as a classical quantity. Such a treatment appears to be justified in case of strong fields, as they are, in particular, generated by lasers. (In fact, this procedure has proved to be very successful in Lamb's famous gas laser theory[1].) Specifically, in the process of light absorption by a two-level atom the physical picture provided by the semiclassical theory is as follows: The field induces an oscillating electric dipole moment, in the sense of a quantum mechanical expectation value, on the atom, and the total energy flow into the atom is given by the work done by the field on that dipole. Note that in this model absorption appears as a continuous process. This description of light absorption is in close correspondence to classical electrodynamics, the main difference, however, being that the amplitude of the induced dipole moment, contrary to that of a harmonic oscillator, can grow, with time, up to a maximum value only (given by the transition matrix element for the electric dipole operator), irrespective of how intense the incident field might be. Clearly, this feature reflects the saturation effect present in a two-level system.

When calculating the energy flow into the atom, along the lines mentioned, one arrives at the result that its maximum value (corresponding to the maximum value of the induced dipole moment) is larger, by orders of magnitude, than the energy flow in the (undisturbed) incident field through the geometric atomic cross section. (A typical example is presented in Sec. III.) From this, one must conclude that an atom has the ability to "suck up" energy from a spatial region that is by far larger than its own volume. One might put the question as to the underlying specific physical mechanism. Actually, an answer is readily given in the framework of classical electrodynamics. An oscillating dipole generates a wave, in any case, the difference between absorption and emission, as a net result, being brought about only by the different phase relations between the incident and the emitted wave. Specifically, in the absorptive case this phase relation gives rise to the effect that the lines of energy flow in the total field are "bent" in a rather large neighborhood of the atom such as to direct the energy flow into the atom. It is the aim of the present paper to give a detailed picture, based on a numerical study, of this bending phenomenon which has been discussed qualitatively already by Fleming[2].

II. SEMICLASSICAL RADIATION THEORY

In the dipole approximation, a radiation field interacts with an atom only via the electric field strength $E(t)$ acting on the atom. We assume the incident field to be an intense monochromatic plane wave which we describe in classical terms. Thus we can write

$$E(t) = E_0 \cos \omega t. \quad (1)$$

For the sake of simplicity, we suppose the field intensity to be so high that the decay of the upper level due to spontaneous emission can be ignored. Then the Schrödinger equation for the probability amplitudes a_1, a_2 for the lower and the upper level, respectively, reads, in the rotating wave approximation (cf. for example, Paul[3])

$$\begin{aligned} i\dot{a}_1 &= \omega_1 a_1 + K e^{i\omega t} a_2, \\ i\dot{a}_2 &= \omega_2 a_2 + K^* e^{-i\omega t} a_1. \end{aligned} \quad (2)$$

Here, ω_1 and ω_2 are the atomic energies, in units of $\hbar$, and K denotes the effective coupling constant

$$K = -\frac{1}{2\hbar} d_{12} E_0, \quad (3)$$

where d_{12} is the transition matrix element for the electric dipole operator, assumed, in what follows, to be a positive number. (Strictly speaking, both the dipole matrix element and the electric field strength are vectors which we assume to coincide in their directions.)

Eqs. (2) are easily solved. Since we are interested in the absorption process, we start, at $t = 0$, from an initial state where the atom is in its lower level ($a_1(0) = 1$, $a_2(0) = 0$). In the resonance case $\omega = \omega_2 - \omega_1$, the expectation value for the electric dipole moment takes the following simple form[3]

$$\langle D(t) \rangle = d(t) \sin \omega t, \quad (4)$$

where the amplitude $d(t)$ is given by

$$d(t) = d_{12} \sin\left(\frac{d_{12} E_0}{\hbar} t\right). \quad (5)$$

One learns from Eq. (5) that the dipole amplitude is a function of time which, under normal conditions, varies slowly in comparison to the light oscillation. In fact, $d(t)$ grows from its initial value zero to its maximum value d_{12} and, afterwards, decreases until it reaches

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zero again. At the end of this cycle, at time

$$T=\frac{\pi\hbar}{d_{12}E_0}, \tag{6}$$

the atom is in the upper level with certainty, i.e., the transition has finished.

It is easily checked that the energy taken up by the atom during this cycle, as given by the work done by the field on the atom, equals that of a single photon, $\hbar\omega$:

$$\int_0^T E(t)\frac{d}{dt}\langle D(t)\rangle dt = E_0 d_{12}\int_0^T \cos\omega t\,\frac{d}{dt}\left[\sin\omega t\cdot\sin\left(\frac{d_{12}E_0}{\hbar}t\right)\right]dt$$

$$=\omega E_0 d_{12}\int_0^T \sin^2\omega t\cdot\sin\left(\frac{d_{12}E_0}{\hbar}t\right)dt\approx\frac{1}{2}\omega E_0 d_{12}\int_0^T\sin\left(\frac{d_{12}E_0}{\hbar}t\right)dt=\hbar\omega. \tag{7}$$

The assumption made at the beginning of this section, that spontaneous emission has no noticeable effect during the period T, requires

$$T\ll\tau, \tag{8}$$

where τ is the mean lifetime of the upper level with respect to spontaneous emission. Since the quantum mechanical treatment of spontaneous emission yields (cf., for example, Heitler[4])

$$\tau^{-1}=\frac{1}{3\pi\varepsilon_0}\frac{\omega^3}{\hbar c^3}d_{12}^2, \tag{9}$$

the inequality (8) can be rewritten as

$$E_0\gg\frac{1}{3\varepsilon_0}\frac{\omega^3}{c^3}d_{12}. \tag{10}$$

For an estimate, we put $d_{12}\simeq r_0 e$, where r_0 is Bohr's radius and e the electronic charge, $\omega\simeq4\times10^{15}$ s^{-1}. Then, the right-hand side in (10) equals 750 V/m which is equivalent to an energy flow, per m^2 and s, of 746 W/m^2.

III. ENERGY FLOW

In what follows, we focus our attention on that stage of the absorption process, where the energy transfer is maximum, corresponding to a maximum value of the amplitude for the induced dipole moment. As has been pointed out in Sec. II, this will happen at time $T/2$, where T is given by Eq. (6). The time-averaged energy flow into the atom, at $t=T/2$, reads

$$F=\frac{1}{2}\omega E_0 d_{12}, \tag{11}$$

while the (time-averaged) energy flux density in the incident wave is

$$\bar{S}=\frac{1}{2}\varepsilon_0 c E_0^2. \tag{12}$$

We can define an effective absorption cross section σ for the atom by requiring the energy flow through σ, $\bar{S}\sigma$, to be equal to F. From Eqs. (11) and (12) we thus find

$$\sigma=\frac{F}{\bar{S}}=\frac{\omega d_{12}}{\varepsilon_0 c E_0}. \tag{13}$$

Inserting here, for example,

$$E_0=(100/4\pi\varepsilon_0)\,\omega^3/c^3\,d_{12},$$

which is consistent with (10), leads to

$$\sigma=\frac{1}{100\pi}\lambda^2, \tag{14}$$

where λ denotes the wavelength of the radiation. In fact, in the optical frequency range considered here, the absorption cross section σ is by far larger than the geometric cross section of the atom.

In this context, it should be mentioned that a similar consideration, applied, however, to an elastically bound, radiation damped electron, yields the result[5] $\sigma=3\lambda^2/(4\pi)$, in case of resonance, for the steady state that is attained in that classical model. While this value is independent of the field intensity, the (maximum) effective cross section for a two-level atom decreases with growing intensity, as becomes obvious from Eq. (13).

As is well known from classical electrodynamics, the physical mechanism that enables a dipole to "suck up" energy from a large volume surrounding it, is provided by a distortion of the incident field due to interference with the wave emerging from the dipole. Since from Eq. (14) a noticeable distortion will be expected to occur in a spatial region of linear dimensions $\lesssim\lambda$ only, and the dipole amplitude is a slowly varying function of time, we may treat the problem as a stationary one. Strictly speaking, we perform a fully classical calculation, considering the induced atomic dipole moment at $t=T/2$, $d_{12}\sin\omega t$, as a given classical dipole oscillation.

According to the well-known formulas first derived by H. Hertz, the dipole field can be described as follows (cf., for example, Sommerfeld[6])

$$\left.\begin{aligned}
E_r^{(D)}&=\frac{\cos\vartheta\, d_{12}k^2}{2\pi\varepsilon_0 r}\left[\frac{1}{kr}\cos(\omega t-kr)+\frac{1}{(kr)^2}\sin(\omega t-kr)\right],\\
E_\vartheta^{(D)}&=-\frac{\sin\vartheta\, d_{12}k^2}{4\pi\varepsilon_0 r}\left[\left(1-\frac{1}{(kr)^2}\right)\sin(\omega t-kr)-\frac{1}{kr}\cos(\omega t-kr)\right],\\
E_\varphi^{(D)}&=0,\\
H_r^{(D)}&=H_\vartheta^{(D)}=0,\\
H_\varphi^{(D)}&=-\frac{\sin\vartheta\, d_{12}ck^2}{4\pi r}\left[\sin(\omega t-kr)-\frac{1}{kr}\cos(\omega t-kr)\right].
\end{aligned}\right\} \tag{15}$$

Here, we have used spherical coordinates r, ϑ, φ, and the dipole located at $r=0$, has been assumed to oscillate along the z axis ($\vartheta=0$). As usual, k stands for the wave number ($k=\omega/c$).

Let the incident field be a linearly polarized plane wave, propagating in the x direction

$$\left.\begin{aligned}
E_x^{(i)}&=E_y^{(i)}=0,\\
E_z^{(i)}&=E_0\cos(\omega t-kx),\\
H_x^{(i)}&=H_z^{(i)}=0,\\
H_y^{(i)}&=-\sqrt{\frac{\varepsilon_0}{\mu_0}}E_0\cos(\omega t-kx),
\end{aligned}\right\} \tag{16}$$

or, in spherical coordinates,

$$\left.\begin{aligned}
E_r^{(i)}&=\cos\theta E_0\cos(\omega t-kx),\\
E_\theta^{(i)}&=-\sin\theta E_0\cos(\omega t-kx),\\
E_\varphi^{(i)}&=0,\\
H_r^{(i)}&=-\sqrt{\frac{\varepsilon_0}{\mu_0}}\sin\theta\sin\varphi E_0\cos(\omega t-kx),\\
H_\theta^{(i)}&=-\sqrt{\frac{\varepsilon_0}{\mu_0}}\cos\theta\sin\varphi E_0\cos(\omega t-kx),\\
H_\varphi^{(i)}&=-\sqrt{\frac{\varepsilon_0}{\mu_0}}\cos\varphi E_0\cos(\omega t-kx).
\end{aligned}\right\} \tag{17}$$

Now, it is a straightforward matter to evaluate from formulas (15) to (17) the Poynting vector **S** for the combined field. According to the assumption (10), the contribution to **S** from the dipole field alone can be disregarded, and we can write, to a good approximation,

$$\mathbf{S} = \mathbf{S}^{(i)} + \mathbf{S}' + \mathbf{S}'', \tag{18}$$

where $\mathbf{S}^{(i)}$ is the Poynting vector for the incident field

$$\mathbf{S}^{(i)} = [\mathbf{E}^{(i)} \times \mathbf{H}^{(i)}], \tag{19}$$

and $\mathbf{S}'$, $\mathbf{S}''$ are the mixed contributions

$$\mathbf{S}' = [\mathbf{E}^{(i)} \times \mathbf{H}^{(D)}], \quad \mathbf{S}'' = [\mathbf{E}^{(D)} \times \mathbf{H}^{(i)}]. \tag{20}$$

After some algebra, we find the different contributions to the time-averaged Poynting vector $\mathbf{S}$ to be given by

$$\left.\begin{aligned} &\bar{S}_x^{(i)} = \frac{1}{2}\sqrt{\frac{\varepsilon_0}{\mu_0}}E_0^2 = \frac{1}{2}c\varepsilon_0 E_0^2,\\ &\bar{S}_y^{(i)} = \bar{S}_z^{(i)} = 0 \end{aligned}\right\} \tag{21}$$

and

$$\left.\begin{aligned} \bar{S}_r' &= -\frac{E_0 d_{12}ck^2\sin^2\theta}{8\pi r}\left[\sin k(r-x) + \frac{1}{kr}\cos k(r-x)\right],\\ \bar{S}_r'' &= -\frac{E_0 d_{12}ck^2\sin\theta\cos\varphi}{8\pi r}\left[\left(1-\frac{1}{(kr)^2}\right)\right.\\ &\qquad\left.\times \sin k(r-x) + \frac{1}{kr}\cos k(r-x)\right],\\ \bar{S}_\theta' &= -\frac{E_0 d_{12}ck^2\sin\theta\cos\theta}{8\pi r}\left[\sin k(r-x) + \frac{1}{kr}\cos k(r-x)\right],\\ \bar{S}_\theta'' &= \frac{E_0 d_{12}ck^2\cos\theta\cos\varphi}{4\pi r}\left[\frac{1}{kr}\cos k(r-x) - \frac{1}{(kr)^2}\sin k(r-x)\right],\\ \bar{S}_\varphi' &= 0,\\ \bar{S}_\varphi'' &= \frac{E_0 d_{12}ck^2}{8\pi r}\sin\varphi\left\{2\cos^2\theta\left[\frac{1}{(kr)^2}\sin k(r-x)\right.\right.\\ &\quad\left. -\frac{1}{kr}\cos k(r-x)\right] + \sin^2\theta\left[\left(1-\frac{1}{(kr)^2}\right)\sin k(r-x) + \right.\\ &\qquad\left.\left. + \frac{1}{kr}\cos k(r-x)\right]\right\}. \end{aligned}\right\} \tag{22}$$

Formulas (21), (22) have been used for a numerical study. Specifically, the energy flux lines in the x, y-plane

$(\theta = \pi/2$

defined by the differential equation

$$\frac{dy}{dx} = \frac{\bar{S}_y}{\bar{S}_x}, \tag{23}$$

and, similarly, those in the x, z-plane have been determined for the case

$$E_0 = (100/4\pi\varepsilon_0)(\omega^3/c^3)\, d_{12},$$

considered already above. The results are depicted in Figs. 1 and 2. In our opinion, they provide some intuitive insight into the absorption mechanism in that they very clearly display the bending of the energy flux lines that enables the atom to "swallow" much more energy than is streaming, in the undisturbed field, through an area that equals its geometrical cross section. Estimating from the figures the effective absorption cross section of the dipole by looking for the positions, in the undisturbed incident field (at the left edge of the figures), of the remotest flux lines which still manage to reach the singularity at $r = 0$, we find it to be in good agreement with the prediction (14). It is especially interesting to learn from the figures that part of the energy, in fact, flows into the atom in backward direction.

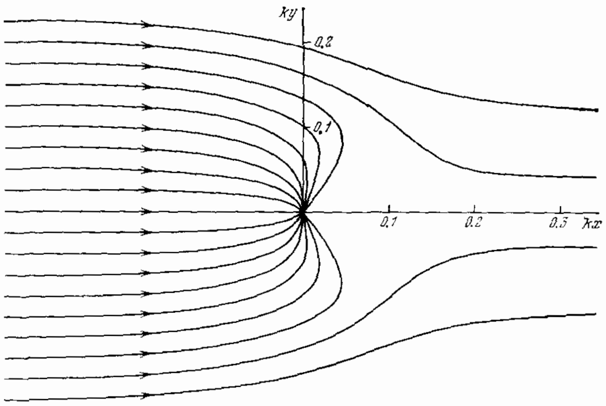

FIG. 1. Energy flux lines in the x, y-plane. The dipole located at $x = y = 0$, oscillates in the z direction. Incident (from the left) is a linearly polarized monochromatic plane wave.

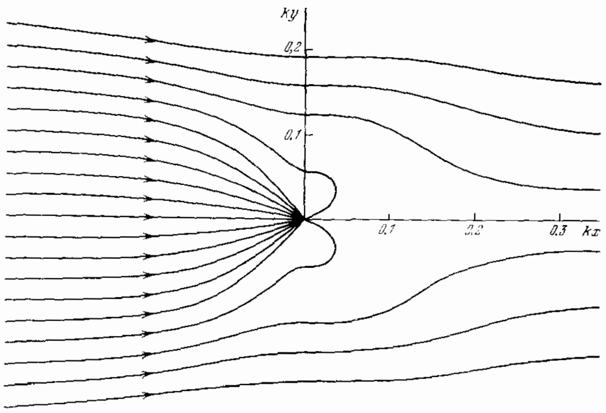

FIG. 2. Energy flux lines in the x, z-plane, for the same physical situation as in Fig. 1.

Actually, our treatment might appear to be rather trivial for people dealing with radio antennas[7]. On the other hand, workers accustomed to the quantum mechanical way of thinking usually visualize an absorption process as "destruction of one photon", and it is perhaps useful to remind them that the corpuscular feature of light represents only one aspect of light, the second one being the wavelike one which predicts a field to emerge from the dipole, even when it absorbs energy from the incident wave. (Peculiarly enough, even M. Planck[8] was not aware of this fact, when estimating the accumulation time for an absorbing atom. Actually, he assumed the maximum energy which an atom can pick up, per s, from the incident field, to be given by the energy flow, in the undisturbed incoming wave, through an area equal to the geometrical cross section of the atom.)

It appears to us that under the conditions studied above, the wave picture is more adequate. An essential assumption, tacitly made in our treatment, is the absence of perturbations from the environment. There is no doubt, of course, that in the opposite case, e.g., when collisions take place rather frequently, an atom may absorb a photon in times very much shorter than the "classical" transition time T. This phenomenon clearly manifests the corpuscular feature of light, and it cannot be accounted for in any classical theory.

Finally, we should like to mention that Figs. 1 and 2, on reversing the direction of the energy flux lines, describe the process of stimulated emission, as viewed from the standpoint of classical electrodynamics.

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Note: H. Paul and R. Fischer, comment on the paper "How can a particle absorb more than the light incident on it" by Craig F. Bohren, Am. J. Phys. **51**, 327 (1983).

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Note. English text kindly supplied by the authors in the form of preprint 82-2 (May 1982).