Anomalous absorption of light by a microscopic particle

G. N. Nikolaev
Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR

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The absorption of light by a microscopic particle near an atom which has an optical transition frequency at resonance with the frequency of the light is analyzed. It is predicted that the energy absorption by the microscopic particle should be increased significantly. The magnitude of the effect depends on the distance between the microscopic particle and the atom and on the temporal conditions of the illumination.

1. The cross section for the absorption of light by an isolated spherical microscopic particle with a radius a much smaller than the wavelength of the incident light, λ ($a/\lambda \ll 1$), is given by the classical formula[1]

$$\sigma_p = 12\frac{a}{\lambdabar}\,\frac{\epsilon''}{|\epsilon+2|^2}\,S, \tag{1}$$

where $\kappa = \lambda/2\pi$, $S = \pi a^2$, and $\epsilon = \epsilon' + i\epsilon''$ is the complex susceptibility of the microscopic particle (we will say simply "particle"). As a rule, σ_p is smaller than the geometric cross section of the particle, S.

On the other hand, we know that the cross section for the resonant interaction of an atom with light is substantially larger: $\sigma_A = 2\pi\gamma/\Gamma\lambdabar^2$, where λ and Γ are the radiative width and total width of the resonant transition of the atom.

In this letter we wish to examine the possibility of a cascade process in which energy is transferred from the light to the atom and then from the atom to the particle.

2. Energy is transferred from the atom excited by the light to the particle through the absorption of the electric field of the atomic dipole induced by the light.

We accordingly consider the auxiliary problem of the absorption of the electric field of a dipole $\mathbf{d}$ by the particle. This dipole is oscillating at a frequency ω and is at a distance $\mathbf{R}$ from the center of the particle.

To calculate the power absorbed in the particle, $\mathscr{P}$, it is convenient to start from the expression[1]

$$\mathscr{P} = -\frac{1}{2}\,\mathrm{Re}\int \mathbf{P}(\mathbf{r})\dot{\mathscr{E}}^*(\mathbf{r})\,d^3r, \tag{2}$$

where the integration is carried out over the volume of the particle, and $\mathbf{P}(\mathbf{r})$ and $\vec{\mathscr{E}}(\mathbf{r})$ are the polarization per unit volume of the particle and the field of the electric dipole at the point $\mathbf{r}$. Equation (2) can be put in the more convenient form

$$\mathcal{P} = -\frac{\omega}{8\pi} \operatorname{Im} \oint (\epsilon - 1)\varphi(\mathbf{r})\vec{\mathcal{E}}^*(\mathbf{r})d\mathbf{S}, \tag{2a}$$

where the integration is now over the surface of the particle, and $\varphi(\mathbf{r})$ is the total electric potential at the surface. For the problem under consideration here, the most interesting situation is that with $R \ll \lambda$. In this case we can use the quasistatic approximation for φ and $\vec{\mathcal{E}}$. The power $\mathcal{P}$ found as a result, plotted as a function of ϵ and a/R, is expressed in terms of hypergeometric functions. The expression for this power simplifies substantially when the distance from the dipole to the surface of the particle is small $(R - a \ll a)(\cos\alpha = \mathbf{dR}/dR)$:

$$\mathcal{P} \approx \frac{\omega|d|^2(1 + \cos^2\alpha)}{16(R - a)^3} \operatorname{Im} \frac{(\epsilon - 1)}{(\epsilon + 1)} . \tag{3}$$

This expression, we might note, is the same as the exact expression for $\mathcal{P}$ in the case in which the particle is replaced by a half-space.

3. The induced dipole arises as a result of three fields: the original field of the light wave, the field of the dipole of the particle which is induced by this wave, and the polarization field of the particle which is produced by the atomic dipole: the "image." We know (Ref. 2, for example) that the latter interaction leads to a shift of the energy levels and a broadening Γ_d of the optical transitions of an atom near material objects. The broadening can be written (Ref. 2, for example)

$$\Gamma_d = 2d_\alpha^{mn} d_\beta^{nm} \operatorname{Im} G_{\alpha\beta}(\mathbf{R}, \mathbf{R}; \omega_{mn})/\hbar, \tag{4}$$

where d_α^{mn} is the matrix element of component α of the dipole moment of the m–n transition, and $G_{\alpha\beta}(\mathbf{R},\mathbf{R};\omega)$ is the field susceptibility:

$$\mathcal{E}_\alpha(\mathbf{r}, \omega) = G_{\alpha\beta}(\mathbf{r}, \mathbf{r}'; \omega) d_\beta(\mathbf{r}', \omega).$$

The quantities $G_{\alpha\beta}$ found for the case at hand are expressed in terms of hypergeometric functions which depend on ϵ and a/R. The expression for $G_{\alpha\beta}$ simplifies substantially when the dipole is close to the surface of the particle:

$$2G_{xx} = 2G_{yy} \approx G_{zz} \approx \frac{1}{4(R - a)^3} \frac{(\epsilon - 1)}{(\epsilon + 1)} \tag{5}$$

(the z axis runs along the axis connecting the atom and the particle).

Substituting these relations into (4), and using the equalities $|d_x^{mn}|^2 = |d_y^{mn}|^2 = |d_z^{mn}|^2 = |d^{mn}|^2/3$ (d^{mn} is the reduced matrix element of the dipole moment of the m–n transition), we find

$$\Gamma_d \approx \frac{|d^{mn}|^2}{3\hbar(R - a)^3} \operatorname{Im}\left(\frac{\epsilon - 1}{\epsilon + 1}\right). \tag{6}$$

The magnitude of the induced atomic dipole depends on the particular conditions under which the system is illuminated. We will consider two cases here: steady-state and pulsed illumination.

4. During steady-state illumination of a two-level atom, the value of $|d|^2$ of the atom is known[3] to behave in a nonmonotonic fashion as a function of the light intensity. At resonance, it reaches a maximum value $|d^{01}|^2/4$ at the saturating value of the field acting on the atom, $|\mathscr{E}_s|^2 = 2\hbar^2\Gamma^2/|d^{01}|^2$, where $\Gamma = \gamma + \Gamma_d$.

The maximum relative efficiency of the energy transfer to the particle in a cascade fashion, $\mathscr{P}$, expressed as a fraction of the energy which is absorbed directly from the light wave by the particle, $\mathscr{P}_p$, can be found from (1), (3), and (6) under these conditions:

$$\eta \equiv \mathscr{P}/\mathscr{P}_p = \frac{3}{64}\,(R/a-1)^3|\epsilon+1|^2|3\epsilon+1|^2/\epsilon''^2. \tag{7}$$

In the derivation of (7) we assumed $\Gamma_d \gg \gamma$ (this is a legitimate assumption when the distance from the atom to the particle is small), and we assumed that the atom-particle system was oriented along the direction of the electric field of the light. For the orthogonal orientation we would have to replace $|3\epsilon+1|$ in (7) by $3/\sqrt{2}$.

From (7) we find a rather unexpected result: The role played by the cascade energy transfer decreases as the atom moves closer to the particle. This result is explained on the basis of a broadening of the transition line as the atom approaches the particle, (6), which hinders the induction of a dipole moment of the atom by the light.

5. We now consider pulsed illumination of a system with a pulse length $\tau_u \ll \Gamma^{-1}$. If the field acting on the atom is a $\pi/2$ pulse[4] with an envelope satisfying the condition $(2|d^{01}|\hbar)\int \mathscr{E}(t)dt = \pi/2$, a maximum oscillatory and damped dipole moment $d = d^{01}\exp(-\Gamma t - i\omega t)$ is induced in the atom after the light pulse passes. In this case, the cascade energy transfer to the particle will evidently be at a maximum. Its value can be found by integrating (3) over t with the expression for d which we just wrote. For a square $\pi/2$ pulse the relative efficiency of the cascade energy transfer to the particle is given by $(\mathbf{d}\|\mathbf{R})$

$$\eta_1 \equiv \int \mathscr{P}dt/\int \mathscr{P}_p dt = \frac{3}{4\pi^2}\left(\frac{\lambda}{a}\right)^3 \frac{|3\epsilon+1|^2}{\epsilon''}\,\gamma\tau_u. \tag{8}$$

We see that despite the small value of $\gamma\tau_u$ (by virtue of our condition that the pulse is short) the quantity η_1 may be quite large because of the large factors $(\lambda/a)^3$ and $|3\epsilon+1|^2/\epsilon''$.

Let us estimate η_1 for a Na particle with a diameter $2a = 100$ Å. We assume that the atom is a Na atom, and we assume that the light has a wavelength $\lambda = 589$ nm and is at resonance with the atom. In this case we find $\eta_1 \simeq 10^6\gamma\tau_u$; i.e., in view of the condition $\gamma\tau_u \ll 1$ we conclude that the efficiency of the cascade energy transfer can reach 10^3–10^4.

In summary, it has been established that the efficiency of the cascade energy transfer from light through a resonant atom to a microscopic particle depends substantially on the illumination conditions. For steady-state illumination, the efficiency of the cascade transfer falls off as the atom moves directly up to the surface of the particle.

For pulsed illumination, the cascade energy transfer to the particle can be efficient by a factor of more than 10^3 than direct absorption of light by the particle.

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