Scattering of Very Intense Electromagnetic Waves*

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The conventional dispersion formula for the scattering cross section of light is extended to include the case of a very high intensity light. The resulting formula predicts an appreciable decrease of the cross section compared to the conventional theory, as the intensity of the light increases. The decrease appears when the energy density of the light becomes comparable to $[\hbar(\omega_0-\omega)/M]^2\epsilon_0$, where ω_0 is the resonance frequency of the scattering medium, ω is the frequency of the light, M is the transition dipole moment, and ϵ_0 is the capacitivity of the vacuum. In a typical case, this means we need about 10^{10} W/cm^2 of the light intensity, which is obtainable with a focused optical maser.

INTRODUCTION

THE scattering of electromagnetic waves by an atom has been investigated theoretically by many people. The classical dispersion formula is discussed by Lorentz,[1] and its quantum-mechanical deviation is obtained by Kramers and Heisenberg,[2] and Waller.[3] All these discussions are conveniently summarized in Heitler's text book.[4]

The classical and the quantum-mechanical formulas are both obtained by assuming that the electromagnetic wave is a small perturbation compared to the atomic energy. The recent invention of masers, however, makes it possible to have a very intense coherent electromagnetic wave. A ruby optical maser is known to produce at least 10^5 W/cm^2 and there is no reason that this should be the upper limit.[5] It is, thus, worthwhile to reinvestigate the problem to see what happens for a very intense electromagnetic wave.

TIME-DEVELOPMENT OPERATOR

The Schrödinger wave equation,

$$i\hbar\partial\Psi/\partial t=H\Psi, \tag{1}$$

can be solved symbolically as

$$\Psi(t)=\exp(-iHt/\hbar)\Psi(0), \tag{2}$$

if the Hamiltonian H does not depend on time explicitly, which is true in the Schrödinger representation. $\Psi(t)$ is the wave function of time t, while $\Psi(0)$ is that at the initial time. The operator $\exp(-iHt/\hbar)$ is called the time-development operator.

The probability of finding a state f at time t starting from another state i is

$$I_{i\to f}(t)=\left|\int\Psi_f^*\Psi_i(t)\,dv\right|^2=\left|\int\Psi_f^*(0)\exp(-iHt/\hbar)\Psi_i(0)\,dv\right|^2, \tag{3}$$

using (2). All radiation process problems can be solved by this formula with

$$H=H_a+H_r+H_i, \tag{4}$$

where H_a is the Hamiltonian of an atom, H_r is that of the radiation field, and H_i gives the interaction between the atom and the radiation field.

Each part of the Hamiltonian is well known[4] and in a representation in which H_a and H_r are orthogonal we have

$$(a\,n_1n_2\cdots n_\lambda\cdots n_\mu\cdots|H_a+H_r|a\,n_1n_2\cdots n_\lambda\cdots n_\mu\cdots)=W_a+\hbar(\omega_1n_1+\omega_2n_2+\cdots+\omega_\lambda n_\lambda+\cdots+\omega_\mu n_\mu+\cdots), \tag{5}$$

where W_a is the energy of the atom in the a state, ω_λ is the frequency of the λth radiation mode, and n_λ is the quantum number of that mode. The zero-point energy of the radiation field is neglected and presumably W_a includes renormalization. In this representation the interaction part of the Hamiltonian has nondiagonal matrix elements. If the wavelengths of the λth and the nth mode are much larger than the dimension of the atom, the following formulas give good approximations to the Hamiltonian matrix:

$$(b\,n_1\cdots n_\mu+1\cdots|H_i|a\,n_1\cdots n_\mu\cdots)=i\omega_{ba}(b|\mathbf{e}_\mu\cdot\mathbf{M}_e|a)[\hbar(n_\mu+1)/2\omega_\mu\epsilon_0V]^{1/2}, \tag{6}$$

$$(an_1\cdots n_\lambda\cdots n_\mu+1\cdots|H_i|an_1\cdots n_\lambda+1\cdots n_\mu\cdots)=(Ze^2/2m)(\hbar/2\epsilon_0V)[(n_\lambda+1)(n_\mu+1)/\omega_\lambda\omega_\mu]^{1/2}\times(\mathbf{e}_\lambda\cdot\mathbf{e}_\mu), \tag{7}$$

where

$$\omega_{ba}=(W_b-W_a)/\hbar. \tag{8}$$

V is the volume of the cavity, ϵ_0 is the capacitivity of vacuum, e and m are charge and mass of the electron,

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[1] H. A. Lorentz, *The Theory of Electrons* (Dover Publications, Inc., New York, 1915).
[2] H. A. Kramers and W. Heisenberg, Z. Physik **31**, 681 (1925).
[3] I. Waller, Z. Physik **51**, 213 (1938).
[4] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1954).
[5] W. Kaiser and C. G. B. Garrett, Phys. Rev. Letters **7**, 229 (1961); P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, *ibid.* **7**, 118 (1961).

respectively, $\mathbf{e}_\lambda$ and $\mathbf{K}_\lambda$ are polarization and propagation vectors of the λth mode, $\mathbf{M}_e$ is the electric dipole moment of the atom, and Z is the number of electrons in the atom.

Suppose we find eigenvalues E_m and eigenfunctions Φ_m of our Hamiltonian H. We can express our initial and final states as

$$\Psi_i=\sum_m(m|i)\Phi_m, \tag{9}$$

$$\Psi_f=\sum_m(m|f)\Phi_m, \tag{10}$$

and the transition $i\rightarrow f$ to be found at time t is, according to (3),

$$I_{i\rightarrow f}(t)=|\sum_m(f|m)(m|i)\exp(-iE_mt/\hbar)|^2, \tag{11}$$

since the Φ's are eigenfunctions of H and are orthonormal to each other. This equation formally solves the transition problems.

CONVENTIONAL SCATTERING FORMULA

Although we solved the problem formally, it is rather difficult to follow this procedure exactly since it is almost impossible to obtain exact eigenvalues of our Hamiltonian. We, therefore, use the perturbation theory to obtain approximate solutions. Since our Hamiltonian has no time dependence, it is the ordinary perturbation theory and not the time-dependent perturbation theory we are using, although we are discussing a time-development problem.

Suppose the initial state is such that the atom is in its ground state a and photon quantum numbers are all zero except for the λth mode. We also assume that the frequency of this λth mode does not satisfy the resonance condition with the atom, or we cannot find any atomic excited state a' for which

$$\omega_\lambda=\omega_{a'a}. \tag{12}$$

If there exists such an atomic excited state, we will have the absorption or the resonance-scattering problem and not the ordinary scattering problem we are considering here.

If the cavity is large compared to the wavelength of the λth mode, there exist other modes whose frequencies are quite close to ω_λ. Let us call one of these modes μ. We see then that our initial state $|a\,00\cdots n0\cdots)$, where only the λth mode is excited to the quantum-number n state, has almost the same energy as another state $|a\,00\cdots n-1\,0\cdots010\cdots)$, where the λth mode is in the $n-1$ state and the μth mode is excited to the quantum-number 1 state. In applying the perturbation theory this degeneracy is to be removed first. Since these two states are connected by the matrix element (7) directly, this part of the Hamiltonian matrix can be written, neglecting all radiation modes except for the λ and μ modes,

$$\begin{bmatrix}(a\,n0|H|a\,n0) & (a\,n0|H|a\,n-11)\\ (a\,n-11|H|a\,n0) & (a\,n-11|H|a\,n-11)\end{bmatrix}, \tag{13}$$

where all matrix elements can be obtained from (5) and (7).

It is easy to see that if we obtain approximate eigenvalues and eigenfunctions from this small matrix (13) and use formula (11), we obtain the Waller term[3] of the scattering probability:

$$\lim_{t\rightarrow\infty} I(a\,n0\rightarrow a\,n-11;t)$$
$$\cong(2\pi/\hbar^2)|(a\,n0|H|a\,n-11)|^2t\delta(\omega_\lambda-\omega_\mu). \tag{14}$$

Excited atomic states can appear through matrix elements (6). The effect of such matrix elements can be taken into account by using a perturbation theory if the photon number is not too large. The Van Vleck transformation,[6] which is a second-order perturbation, is suitable in our calculation. If we modify each matrix element of (13) in this way we obtain the Kramers-Heisenberg dispersion formula[2] with the Waller term:

$$\operatorname*{Lim}_{t\rightarrow\infty} I(a\,n0\rightarrow a\,n-11;t)$$
$$\cong(2\pi/\hbar^2)n(\omega/\epsilon_0V)^2\{\sum_b(b|\mathbf{e}_\lambda\cdot\mathbf{M}_e|a)(a|\mathbf{e}_\mu\cdot\mathbf{M}_e|b)$$
$$\times[(\omega_{ba}-\omega)^{-1}+(\omega_{ba}+\omega)^{-1}]\}^2t\delta(\omega_\lambda'-\omega_\mu'), \tag{15}$$

where

$$\omega\cong\omega_\lambda \quad \text{or} \quad \omega_\mu,$$

while

$$\omega_\lambda'=\omega_\lambda+\sum_b(\omega/2\hbar\epsilon_0V)|(b|\mathbf{e}_\lambda\cdot\mathbf{M}_e|a)|^2$$
$$\times[(\omega_{ba}-\omega)^{-1}+(\omega_{ba}+\omega)^{-1}], \tag{16}$$

and a similar expression for ω_μ'.

The energy conservation rule,

$$\omega_\lambda'=\omega_\mu', \tag{17}$$

can be reasonably interpreted when we observe that the added term in (16) is nothing but α_λ/V, where α_λ is the polarizability of the atom for the λth mode of the field. It is easy to see that if we have a system of N atoms this added term will be $N\alpha_\lambda/V$, so that our conservation rule (17) means that ω_x (index of refraction) should be conserved for the scattering process.

SCATTERING OF INTENSE ELECTROMAGNETIC WAVE

If the photon number n of a mode is very large, it can reach a point where

$$\omega_{ba}(b|\mathbf{e}\cdot\mathbf{M}_e|a)[\hbar n/2\omega\epsilon_0V]^{1/2}\cong h|\omega_{ba}-\omega|. \tag{18}$$

In this case the conventional formula (15), which is essentially the second-order perturbation term, is not a good approximation to take into account the atomic virtual excitation. In the case of the optical region this can happen if the beam intensity is 10^{10} W/cm^2 or more. This is not impossible for a focused maser beam.

[6] J. H. Van Vleck, Phys. Rev. **33**, 467 (1929).

In the microwave region the same situation can appear at 10^5 W/cm^2.

The initial state of the scattering $|a\,n\,0)$ is connected to states $|b\,n-1\,0)$ and $|b\,n+1\,0)$ through the virtual atomic excitation, while the final state $|a\,n-1\,1)$ is connected to $|b\,n-2\,1)$ and $|b\,n\,1)$ states in the same way. The matrix for these six states is obtained by (6) and (7) as

$$\begin{bmatrix} 0 & A'n^{1/2} & A'(n+1)^{1/2} & Bn^{1/2} & 0 & C' \\ & \hbar(\omega_{ba}-\omega_\lambda) & 0 & C' & B(n-1)^{1/2} & Bn^{1/2} \\ & & \hbar(\omega_{ba}+\omega_\lambda) & 0 & 0 & B(n+1)^{1/2} \\ & & & \hbar(\omega_\mu-\omega_\lambda) & A'(n-1)^{1/2} & A'n^{1/2} \\ & & & & \hbar(\omega_{ba}-2\omega_\lambda+\omega_\mu) & 0 \\ & \text{Hermitian} & & & & \hbar(\omega_{ba}+\omega_\mu) \end{bmatrix}, \tag{19}$$

where

$$\begin{aligned} A' &= i\omega_{ba}(b|\mathbf{e}_\lambda\cdot\mathbf{M}_e|a)(\hbar/2\omega_\lambda\epsilon_0 V)^{1/2}, \\ C' &= i\omega_{ba}(b|\mathbf{e}_\mu\cdot\mathbf{M}_e|a)(\hbar/2\omega_\mu\epsilon_0 V)^{1/2}, \\ B &= -(Ze^2/2m)(\hbar/2\omega\epsilon_0 V)(\mathbf{e}_\lambda\cdot\mathbf{e}_\mu), \end{aligned} \tag{20}$$

and all diagonal elements of the A^2 term in the Hamiltonian are neglected.

Of course, there exist many other atomic states beside a and b states, but in most atoms the matrix element A or C is very large between the ground state and the first excited state compared to all other combinations. In the case of the hydrogen atom, for example,[7] if a is the $1s$ state, we can take a $2p$ state for b and neglect all others, since A or C between $1s$ and $2p$ is more than 5 times as large as the sum of matrix elements between $1s$ and all bound states except $2p$.

We assume that the photon number n is very large. Since A and C are of the same order of magnitude, we see that matrix elements with A in it are much more important than those with C in (19). We know that B terms in (19) are also unimportant. Actually, Power and Zienau[8] showed that as long as we are interested in the limit $t\to\infty$, the result will not change by neglecting B terms and replacing ω_{ba} by ω_λ and ω_μ in A and C, respectively. The validity of this statement is seen in the conventional formula (15). Since Power and Zienau's scheme will simplify our calculation we will take that in the rest of this paper and define

$$\begin{aligned} A &= i\omega_\lambda(b|\mathbf{e}_\lambda\cdot\mathbf{M}_e|a)(\hbar/2\omega_\lambda\epsilon_0 V)^{1/2}, \\ C &= i\omega_\mu(b|\mathbf{e}_\mu\cdot\mathbf{M}_e|a)(\hbar/2\omega_\mu\epsilon_0 V)^{1/2}. \end{aligned} \tag{21}$$

It is clear that the first approximation for our eigenvalues of (19) can be obtained by diagonalizing the A terms, namely, diagonalizing each 3 by 3 matrix.

In many cases we have

$$\omega_{ba}\gg\omega_\lambda,\quad \omega_\mu. \tag{22}$$

In such case of off-resonance the diagonalization of the first 3 by 3 matrix made by $|a\,n\,0)$, $|b\,n-1\,0)$, and $|b\,n-1\,0)$ gives

$$E_\pm^{(1)} = \hbar\omega_{ba}/2 \pm X^2, \tag{23}$$

$$E_0^{(1)} = \hbar\omega_{ba}, \tag{24}$$

$$\psi_\pm^{(1)} = \xi_\pm|a\,n\,0) + (\eta_\pm/\sqrt{2})[|b\,n-1\,0) + |b\,n+1\,0)], \tag{25}$$

$$\psi_0^{(1)} = 2^{-1/2}[|b\,n-1\,0) - |b\,n+1\,0)], \tag{26}$$

where ξ, η are transformation coefficients given by

$$\xi_\pm = An^{1/2}/XY_\pm, \tag{27}$$

$$\eta_\pm = \pm|\xi_\mp| = \pm Y_\pm/X\sqrt{2}, \tag{28}$$

where

$$X = [2|A|^2 n + \hbar^2\omega_{ba}^2/4]^{1/4}, \tag{29}$$

$$Y_\pm = [X^2 \mp \hbar\omega_{ba}/2]^{1/2}. \tag{30}$$

The diagonalization of the second 3 by 3 matrix gives

$$E_\pm^{(2)} = \hbar(\omega_\mu-\omega_\lambda) + \hbar\omega_{ba}/2 \pm X^2, \tag{31}$$

$$E_0^{(2)} = \hbar(\omega_\mu-\omega_\lambda+\omega_{ba}), \tag{32}$$

$$\psi_\pm^{(2)} = \xi_\pm|a\,n-1\,1) + (\eta_\pm/\sqrt{2})[|b\,n-2\,1) + |b\,n\,1)], \tag{33}$$

$$\psi_0^{(2)} = 2^{-1/2}[|b\,n-2\,1) - |b\,n\,1)]. \tag{34}$$

It is sometimes important to notice that all coefficients in the second case are slightly different from the first case, namely, $n-1$ should be used instead of n. In our present calculation, however, we can neglect such small differences.

We see that out original matrix (19) is now transformed to

$$\begin{bmatrix} E_0^{(1)} & 0 & 0 & \xi_+C/\sqrt{2} & \xi_-C/\sqrt{2} & 0 \\ & E_+^{(1)} & 0 & 2^{1/2}\xi_+\eta_+C\sqrt{2} & \xi_+\eta_-C & -\xi_+C/\sqrt{2} \\ & & E_-^{(1)} & 2^{1/2}\xi_-\eta_+C\sqrt{2} & \xi_-\eta_-C & -\xi_-C/\sqrt{2} \\ & & & E_+^{(2)} & 0 & 0 \\ & \text{Hermitian} & & & E_-^{(2)} & 0 \\ & & & & & E_0^{(2)} \end{bmatrix}. \tag{35}$$

When $\omega_\mu\cong\omega_\lambda$ we see that

$$\begin{aligned} E_+^{(1)} &\cong E_+^{(2)}, \\ E_-^{(1)} &\cong E_-^{(2)}; \end{aligned} \tag{36}$$

[7] See H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic Press Inc., New York, 1957); and E. V. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, New York, 1953).

[8] E. A. Power and S. Zienau, Phil. Trans. Roy. Soc. London **251**, 427 (1959).

thus

$$\tfrac{1}{2}[E_+^{(1)}+E_+^{(2)}]\pm[\hbar^2(\omega_\lambda-\omega_\mu)^2/4+2|\xi_+\eta_+C|^2]^{1/2}, \quad (37)$$

$$: \varphi_\pm=\xi_\pm'\psi_+^{(1)}+\eta_\pm'\psi_+^{(2)}, \quad (38)$$

$$\tfrac{1}{2}[E_-^{(1)}+E_-^{(2)}]\pm[\hbar^2(\omega_\lambda-\omega_\mu)^2/4+2|\xi_-\eta_-C|2]^{1/2}, \quad (39)$$

$$: \varphi_\pm'=\xi_\pm'\psi_-^{(1)}+\eta_\pm'\psi_-^{(2)} \quad (40)$$

will be the improved eigenvalues and eigenfunctions.

Since

$$|a\,n\,0)$$
$$=-\xi_+\psi_+^{(1)}-\xi_-\psi_-^{(1)}$$
$$=-\xi_+(\xi_+'\,\varphi_+'+\xi_-'\,\varphi_-)-\xi_-(\xi_+'\,\varphi_+'+\xi_-'\,\varphi_-'), \quad (41)$$

$$|a\,n-1\,1)$$
$$=-\xi_+(\eta_+'\varphi_+'+\eta_-'\varphi_-)-\xi_-(\eta_+'\,\varphi_+'+\eta_-'\,\varphi_-'), \quad (42)$$

the scattering probability is

$$\lim_{t\to\infty}|(a\,n-11|\exp(-iHt/\hbar)|a\,n0)|^2/t$$
$$=(2\pi/\hbar^2)|\xi_+|^4 2|\xi_+\eta_+C|^2\delta[(E_+^{(1)}-E_+^{(2)})/\hbar]$$
$$+(2\pi/\hbar^2)|\xi_-|^4 2|\xi_-\eta_-C|^2\delta[E_-^{(1)}-E_-^{(2)})/\hbar]$$
$$\cong(2\pi/\hbar^2)|A|^6|C|^2n^3(2|A|^2n+\hbar^2\omega_{ba}^2/4)^{-2}$$
$$\times[Y_+^{-4}+Y_-^{-4}]\delta(\omega_\lambda-\omega_\mu)$$
$$=(2\pi/\hbar^2)|AC|^2n(|A|^2n+\hbar^2\omega_{ba}^2/4)/$$
$$\times(2|A|^2n+\hbar^2\omega_{ba}^2/4)^2\delta(\omega_\lambda-\omega_\mu). \quad (43)$$

When the photon number n is small this formula reduces to

$$(2\pi/\hbar^2)4|AC|^2n/(\hbar\omega_{ba})^2\delta(\omega_\lambda-\omega_\mu), \quad (44)$$

which agrees with the conventional scattering formula in the nonresonant region.

When n is very large, however, our formula (43) predicts that the scattering cross section will decrease considerably from the conventional value. The calculated scattering cross sections are given in Fig. 1.

It is easy to investigate another limiting case, namely,

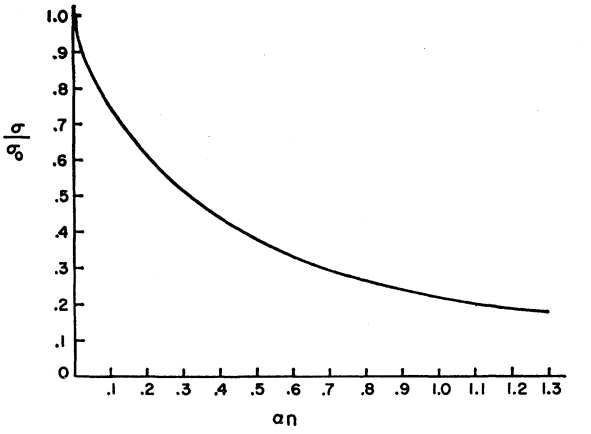

FIG. 1. Scattering cross section as a function of photon number n. σ_0 is the conventional scattering cross section. $\alpha=4|A|^2/\hbar^2\omega_{ba}^2$ for the nonresonant case, while $\alpha=2|A|^2/\hbar^2(\omega_{ba}-\omega)^2$ for the nearly resonant case.

the nearly resonant case, where

$$\omega_{ba}\cong\omega_\lambda, \quad \omega_\mu. \quad (45)$$

As we can see from (19), the effect of $|b\,n+1\,0)$ and $|b\,n\,1)$ states are negligible compared to those of $|b\,n-1\,0)$ and $|b\,n-2\,1)$ states, in the nearly resonant case. Under such circumstances the calculation can be done easily and we obtain the final result

$$\lim|(a\,n-1\,1|\exp(-iHt/\hbar)|a\,n\,0)|^2/t$$
$$=(2\pi/\hbar^2)|AC|^2n[2|A|^2n+\hbar^2(\omega_{ba}-\omega)^2]$$
$$\times[4|A|^2n+\hbar^2(\omega_{ba}-\omega)^2]^{-2}\delta(\omega_\mu-\omega_\lambda). \quad (46)$$

We see that this formula also reduces to the conventional one when n is small, but predicts a considerable decrease of the scattering cross section when n is large. The calculated results for the nearly resonant case is also shown in Fig. 1.

A photon beam of 10^5 W/cm^2 means that $hn\omega/V$ is about 3 J/m^3. For a typical case this value gives the ratio $|A|^2n/\hbar^2\omega_{ba}^2$ to be about 10^{-10}. The anomaly calculated here may be detectable when a photon beam of 10^{10} W/cm^2 is used. The anomaly also appears in the index of refraction. The latter will be discussed in a separate paper.