MULTIPHONON PROCESSES ASSOCIATED WITH THE RESONANT SCATTERING OF LIGHT BY ATOMS

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The effect of a strong electromagnetic field on the process of resonant scattering of light by atoms is examined by the method of Green's functions. A new type of resonant scattering is predicted at frequencies at which the atom would scatter light weakly in the absence of electromagnetic radiation.

RECENTLY the multiphoton absorption of light by atoms and by condensed media[1–6] has been investigated in detail. It is clear, however, that the quasi-energy character of the spectrum of an atom which is interacting with intense radiation[7] may appear not only in absorption processes but also in processes of light scattering. In particular, one would expect the well-investigated resonant scattering of light by atoms to change in the presence of laser radiation due to the appearance of a new type of resonances. The goal of the present communication is the investigation of precisely this range of problems.

Let us use the well-known formula expressing the cross-section for the scattering of light by an atom in terms of the Green's function (see [8], formula (27.1.3)). It is obvious that in the case when the atom is located in a field of intense laser radiation, the Green's function of the free atom should be replaced by the Green's function $G(x_1, x_2)$ for an atom in the field of a strong electromagnetic wave.[9]

Such a substitution corresponds to the diagram shown in Fig. 1 ($A(x)$ is the vector potential of the scattered light, $\Psi_{1s}(x)$ is the wave function of the atom in its ground state ($x = \mathbf{r}, t$)). For simplicity we regard it as unperturbed by the laser radiation:[1)] $\Psi_{1s}(x) = \Psi^0_{1s}(\mathbf{r}) \exp\{iE^0_{1s}t\}$.

First let us consider the resonant scattering of light by the hydrogen atom in its ground state for frequencies Ω_0 which are close to the eigenfrequency $\Omega_{1s\to 2p} = E^0_{2p} - E^0_{1s}$.

In the resonance approximation for the scattering process, the degenerate level (2s, 2p) gives the "major" contribution to the probability. The wave functions of this level in the field of an intense electromagnetic wave can be found exactly if we neglect their mixing with the wave functions of the other levels, i.e., if we confine our attention to a three-level model. In the dipole approximation the correct functions of the excited (2s, 2p) state are written in the form

$$\Psi_1(\mathbf{r}, t) = 2^{-1/2}(\psi_{2p}^0(\mathbf{r}) + \psi_{2s}^0(\mathbf{r}))\exp\{-i(E_{2p}^0 t + \rho_1 \sin\omega t)\},$$
$$\Psi_2(\mathbf{r}, t) = 2^{-1/2}(\psi_{2p}^0(\mathbf{r}) - \psi_{2s}^0(\mathbf{r}))\exp\{-i(E_{2p}^0 t - \rho_1 \sin\omega t)\}, \qquad (1)$$

where $\rho_1 = v_{2p,2s}/\omega$ and $V_{2p,2s} = e_0 F z_{2p,2s}$ (F denotes the amplitude of the field intensity of the laser beam of frequency ω, z is the polarization, and $z_{2p,2s}$ denotes the matrix element of the coordinate).

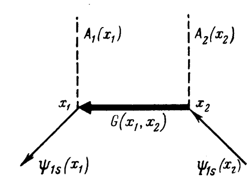

FIG. 1

Therefore, in the resonance approximation the Green's function of the problem takes the form

$$G(\mathbf{r}, t; \mathbf{r}_1, t_1) = \Theta(t - t_1)\sum_{n=1}^{2}\Psi_n(\mathbf{r}, t)\Psi_n^*(\mathbf{r}_1, t_1) + g(\mathbf{r}, t; \mathbf{r}_1, t_1),$$
$$\Theta(t - t_1) = \begin{cases} 1, & t > t_1, \\ 0, & t < t. \end{cases} \qquad (2)$$

The contribution to the Green's function resulting from the remaining part of the spectrum of the atom, which is interacting with the radiation, is denoted by $g(\mathbf{r}, t; \mathbf{r}_1, t_1)$.

We obtain the expression for the scattering cross section by writing down the contribution from the diagram shown in Fig. 1. It is also necessary to use the expansion

$$\exp\{i\rho_1 \sin\omega t\} = \sum_{n=-\infty}^{+\infty} J_n(\rho_1)\exp\{in\omega t\},$$

where $J_n(\rho_1)$ is the Bessel function of n-th order. We find

$$d\sigma = \sum_{n=-\infty}^{+\infty} d\sigma_n,$$

$$d\sigma_n = \Omega_0(\Omega_0 + 2n\omega)^3 \left|\sum_{k=-\infty}^{+\infty}\frac{\langle 1s|\mathbf{Q}\mathbf{e}_2|2p\rangle\langle 2p|\mathbf{Q}\mathbf{e}_1|1s\rangle J_k(\rho_1)J_{2n+k}(\rho_1)}{E_{1s}^0 - E_{2p}^0 + \Omega_0 - k\omega + i\gamma}\right|^2 do_2, \qquad (3)$$

Here **Q** is the dipole moment of the atom, $\mathbf{e}_1$ and $\mathbf{e}_2$ are the unit polarization vectors of the incident and scattered light, and $d\sigma_2$ is the infinitesimal element of solid angle for the scattered light. The attenuation constant γ of the excited state is introduced phenomenologically.

In the absence of laser radiation ($\rho_1 = 0$) the only term which does not vanish will be the term with $n = k = 0$ ($J_0(0) = 1$), and formula (3) goes over into the well-known expression for the resonant scattering of light by the hydrogen atom.

[1)]In this work the system of units in which $\hbar = c = 1$ is adopted.

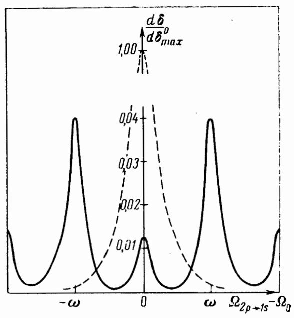

FIG. 2

Let us also give the formula for the cross section for light scattering by an atom with a single optical electron. Let us calculate beforehand the Green's function for this case. One can represent the solution of the time-dependent Schrödinger equation according to method [10] in the form

$$\Psi_n(\mathbf{r}, t) = \psi_n^0(\mathbf{r}) \exp\{-i(E_n^0 t + \alpha_n(t))\},$$
$$\alpha_n(t) = 2\omega\rho_n t + \rho_n \sin 2\omega t - i\bar{\rho}_n \cos 2\omega t,$$
$$\rho_n = \sum_{\substack{m \\ (m\neq n)}} \frac{V_{mn}V_{nm}}{\Omega_{mn}^2 - \omega^2}\frac{\Omega_{nm}}{\omega}; \qquad \bar{\rho}_n = \sum_{\substack{m \\ (m\neq n)}} \frac{V_{mn}V_{nm}}{\Omega_{mn}^2 - \omega^2}. \tag{4}$$

Thus, the expression for the Green's function takes the form

$$G(\mathbf{r}, t; \mathbf{r}_1, t_1) = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} dx \sum_n \psi_n^0(\mathbf{r})\psi_n^{0*}(\mathbf{r}_1)\cdot$$
$$\times \sum_{k,p=-\infty}^{+\infty} \frac{J_k(\rho_n)J_{k+p}(\rho_n)}{(E_n^0 + 2\omega\rho_n + 2p\omega)(1 - i0) + x} e^{-2ik\omega t} e^{ix(t-t_1)}. \tag{5}$$

Consequently the cross section for the scattering of light is determined by the formula

$$d\sigma = \sum_{n=-\infty}^{+\infty} d\sigma_n,$$
$$d\sigma_n = \Omega_0(\Omega_0 + 2n\omega)^3\cdot$$
$$\times \left|\sum_m \sum_{k=-\infty}^{+\infty} \frac{\langle 1s|\mathbf{Qe}_2|m\rangle\langle m|\mathbf{Qe}_1|1s\rangle J_k(\rho_m)J_{n+k}(\rho_m)}{E_{1s}^0 - E_m^0 + \Omega_0 - 2\omega\rho_m - 2k\omega + i\gamma}\right|^2 do_2. \tag{6}$$

For simplicity, formulas (5) and (6) are written down in the approximation $\bar{\rho}_n \ll 1$. (For the scattering of light whose frequency is close to the energy difference between the ground and the first excited states of the atom, the approximation $\bar{\rho}_n \sim 0$ is well satisfied.)

Let us clarify the specific characteristics which the multiphoton processes introduce into the elementary process of light scattering by an atom. As follows from formulas (3) and (6), the scattered light consists of a set of "harmonics" with frequencies $\Omega_n = \Omega_0 + 2n\omega$; $n = 0, \pm 1, \pm 2, \ldots$. The intensities of the harmonics are determined by the Bessel functions and decrease sharply with increase in the number of the harmonic. Taking account of the influence of the laser radiation on the zero harmonic, which corresponds to "Rayleigh" scattering of light by the atom, is of special interest. It is not difficult to show that in this case in the presence of laser radiation the cross section $d\sigma_r$ for resonant scattering of light can be expressed in terms of the cross section $d\sigma_r^0$ for resonant scattering of light by a free atom according to the formula

$$d\sigma_r \approx d\sigma_r^0 J_0^4(\rho). \tag{7}$$

Here $\rho = \rho_1$ in the case of formula (3) and $\rho = \rho_{2p}$ in the case of formula (2).

Therefore, an abrupt suppression of the zero harmonic of the resonant scattering should be observed in connection with those intensities of the laser radiation for which $\rho \sim 1$. (For example, for a neodymium laser with quanta of energy 1.17 eV and for field intensities of 10^7 to 10^8 V/cm, the parameter $\rho \sim 1$. In the case of a CO_2 laser with quanta of energy 0.12 eV the condition $\rho \sim 1$ is already achieved for fields of 10^5 V/cm.)

Another interesting consequence of formulas (3)–(6) is the prediction of an anomalously large scattering of light by an atom at frequencies for which the atom would essentially not scatter any light at all in the absence of the laser radiation.

These frequencies are found from the condition that the frequency of the external scattered light coincide with the quasi-energy levels of an atom which is interacting with intense laser radiation. The resonance conditions have the following form: for scattering by the hydrogen atom $\Omega_r = \Omega_{2p\to 1s} - r\omega$ where $r = 0, \pm 1, \pm 2, \ldots$; for scattering by an atom with one optical electron $\Omega_r = \Omega_{2p\to 1s} - 2(r - \rho_{2p})\omega$ where $r = 0, \pm 1, \pm 2, \ldots$.

The dependence of the resonant scattering of light by the hydrogen atom on the frequency of the incident light in the absence of laser radiation is graphically shown in Fig. 2 (by the dashed curve) and also the frequency dependence of the resonant scattering in the presence of laser radiation (the solid curve) is shown, both in units of the maximum cross section $d\sigma^0_{max}$ for the resonant scattering of light by a free atom. The numerical calculations were carried out for the value $\rho_1 = 1.8$.

In conclusion we note that the relationships investigated here should also exist for the scattering of charged particles by an atom in the field of a strong electromagnetic wave; it is proposed to investigate this case separately.

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