

CONVERSION OF THE VISIBLE COHERENT RADIATION INTO THE INFRARED ONE

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Scattering of external electromagnetic radiation is considered by an example of hydrogen-like atom participating in an oscillation process based on the formalism of effective wave functions describing the mixed quantum states. It is shown that the additional dipole moment caused by the influence of neighbor atoms results in appearance, in the scattered radiation, of the low frequency components with the frequency of atom oscillations in a thermostat.

One of the main problems of linear and nonlinear optics is the problem on interaction between the radiation and the matter. There are some achievements here, especially in the nonlinear optics. The methods of frequency conversion of the external radiation by crystals and molecules are well studied both theoretically and experimentally. However, the historical background shows that the main attention was paid to the problem of the radiation frequency multiplication. The problem on downshifting the radiation frequency due to nonlinear interaction is significantly less studied. At the same time, the laws of conversion of visible radiation into the IR one are important especially for applied purposes because they determine the energy budget of complex systems.

This paper is devoted to the study of a possible mechanism of downshifting visible radiation to IR by an ensemble of atoms or molecules.

The initial statement is an obvious physical assumption that the kinetic degrees of freedom of atoms (for example, oscillations) in molecules or crystals should affect the scattering of external radiation, especially, if one takes into account that, in the general case, such oscillations can be synchronous for a group of atoms in the lattice sites of a crystal or in the neighbor molecules. This synchronization can occur due to the photon exchange (when an acoustic wave propagates through the medium) or under the effect of light with the frequency close to the natural oscillation frequencies¹ (as will be shown below, such electromagnetic radiation appears spontaneously at the presence of an acoustic wave). The study of the effect of such a kind of cooperative phenomena on the scattering spectrum is the subject of this paper.

To find the conditions for generation of the low frequency electromagnetic waves in the medium, it is necessary to solve two inter-related problems. First, one should find the response of one atom to the external electromagnetic field in the case when this atom takes part in some collective motion. And second, one should find the conditions that are necessary for generation of electromagnetic field at Raman frequencies by an ensemble of such atoms.

Let us consider an ensemble of hydrogen-like atoms oscillating in some thermostat as an example of

solving these problems. Such a choice is convenient for two reasons. On the one hand, if refer to the ideas and final formulas of the Hartree-Fock method,² such an idealization of the problem quite well approximates the state of real atoms that are the elements of some ensemble. On the other hand, the choice of a hydrogen-like atom as a model for calculations makes it possible to derive final mathematical expressions that do not contain unknown variables, i.e. to derive the exact formulas.

To calculate the microscopic induced dipole moment of the hydrogen-like atom oscillating in a thermostat, let us use the wave equation for the auxiliary wave function averaged over the thermostat effect:

$$i\hbar \frac{\partial \Psi}{\partial t} = \frac{1}{1+i\alpha} \left[\frac{1}{2m} \left(\hat{\mathbf{P}} - \frac{e}{c} \mathbf{A} - (1-i\alpha) m \mathbf{V} \right)^2 + \kappa \right] \Psi + e \left(\phi - (\mathbf{A} \cdot \mathbf{V}) / 2c - (1+i\alpha) \frac{m \mathbf{V}^2}{8e} \right) \Psi, \quad (1)$$

where α and κ are the positive parameters that take into account the collisional perturbation of an atom (they are assumed to be empirical values); \mathbf{V} is the atom velocity in the thermostat; $\hat{\mathbf{P}}$ is the pulse operator; \mathbf{A} and ϕ are the vector and scalar potentials, respectively; e and m are the electron charge and mass.

Equation (1) is constructed by the method of integrals over Feynman trajectories³ as a result of averaging the Feynman propagator over the probability of occurrence of a random trajectory. One of the ways to construct this equation has been described in Ref. 4.

The total wave function describing the behavior of a hydrogen-like atom in the thermostat is connected with the solution of Eq. (1) by the relationship⁵:

$$\Psi = \Psi / \langle \Psi | \Psi \rangle^{1/2}. \quad (2)$$

The fact that both the vector-potential of the external electromagnetic field and the drift velocity of the atom as a whole in the thermostat are included in Eq. (1) allows us to describe the state of the quantum system in the case of interference of these external factors perturbing the atom.

When solving Eq. (1) let us take into account that, in comparison with the electronic states of atoms and molecules, oscillations make-up a slowly changing dynamic subsystem. This fact makes it possible to find the wave function satisfying Eq. (1) by means of the iteration method considering the oscillation effect in the adiabatic approximation.

Let us make some assumptions related to the electromagnetic field whose vector-potential is included in Eq. (1).

Let us assume that the field affecting the atom can be represented in the form of superposition of a strong plane-polarized monochromatic radiation of the frequency close to the frequency of the quantum transition from the second level to the first one (we assume that the radiation is generated by an external source), and quite a weak radiation of all other frequencies. The difference between the frequency ω of radiation from an external source and the frequency ω_{21} of the quantum transition ($\varepsilon = \omega - \omega_{21}$) satisfies the condition

$$|\varepsilon| \ll \omega. \quad (3)$$

in accordance with the assumptions accepted.

If to pass from the vector-potential to the electric field intensity in Eq. (1) and neglect the terms that make small corrections to the solution, one can write the following expression for the hydrogen-like atom in the dipole approximation:

$$i\hbar \frac{\partial \psi}{\partial t} = \frac{1}{1+i\alpha} \left(\frac{1}{2m} \hat{\mathbf{p}}^2 \right) \psi - \frac{Z e^2}{r} \psi + (\mathbf{E} \mathbf{d}) \psi - \frac{m}{2e} (1-i\alpha) \left(\frac{\partial \mathbf{V}}{\partial t} \mathbf{d} \right) \psi - \frac{m}{8} (1+i\alpha) \mathbf{V}^2 \psi. \quad (4)$$

Here \mathbf{d} is the atomic dipole moment; Z is the nucleus charge multiplicity; r is the radius-vector length.

If to apply the adiabatic approximation to solving Eq. (4), then, turning the Z -axis of the local coordinate system connected with the atom along the direction of oscillations of the vector \mathbf{E} of the external field, it is easy to obtain the following expression for the auxiliary wave function:

$$\psi(\mathbf{r}, t) = \exp \left[-\frac{i}{\hbar} \frac{\kappa}{1+i\alpha} t + \frac{i}{\hbar} \frac{e}{c} \left(\left(\mathbf{A} + \frac{c}{e} (1-i\alpha) \frac{m\mathbf{V}}{2} \right) \cdot \mathbf{r} \right) + \frac{im}{8\hbar} (1+i\alpha) \int \mathbf{V}^2 dt \right] \left[b_1(t) \exp \left(-\frac{i}{\hbar} (1+i\alpha) E_1 t \right) \psi_1(\mathbf{r}) + (b_2(t) \psi_2(\mathbf{r}) + b_3(t) \psi_3(\mathbf{r})) \exp \left(-\frac{i}{\hbar} (1+i\alpha) E_2 t \right) \right], \quad (5)$$

where E_i are the eigenvalues of the energy of quantum states of the hydrogen-like atom, $\psi_i(\mathbf{r})$ are the corresponding eigenfunctions written⁶ for the atom experiencing the collisional disturbance (it is assumed that $\psi_1 = \psi_{100}$; $\psi_2 = \psi_{200}$; $\psi_3 = \psi_{210}$).

Coefficients $b_i(t)$ included in Eq. (5) are approximately equal to:

$$b_1(t) = A_1 \exp(\beta_1 t) + A_2 \exp(\beta_2 t); \quad (6)$$

$$b_3(t) = B_1 \exp(-\beta_2 t) + B_2 \exp(-\beta_1 t); \quad (7)$$

$$b_2(t) = -\frac{3 a_0 m \omega_0 V_z}{4 \hbar Z} B_1 \exp(-\beta_2 t) \times \left[\frac{\exp(i \omega_0 t - i f_0)}{i \omega_0 - \beta_2} + \frac{\exp(-i \omega_0 t + i f_0)}{i \omega_0 + \beta_2} \right] + B_2 \exp(-\beta_1 t) \left[\frac{\exp(i \omega_0 t - i f_0)}{i \omega_0 - \beta_1} + \frac{\exp(-i \omega_0 t + i f_0)}{i \omega_0 + \beta_1} \right] + c. \quad (8)$$

There are the following designations in Eqs. (6)–(8): α_0 is the Bohr radius; A_i , B_i , and Γ are the constants determined by the initial conditions, some of them are related to each other:

$$B_i = i e |\mathbf{E}_0| a_0 2^{13/2} / (3^5 Z \hbar \beta_{2/i}) \exp(i(\mathbf{K} \cdot \mathbf{R})) A_i. \quad (9)$$

The values V_z , ω_0 , and f_0 correspond to the amplitude, frequency, and initial phase of the low-frequency oscillations of the atom in the thermostat (according to the selection rules, only z -component of the amplitude is taken into account); β_i are the constants that can be estimated by the following formulas:

$$\beta_1 = i \left(\varepsilon + \frac{2^{14} e^2 |\mathbf{E}_0|^2 a_0^2}{9^5 \varepsilon \hbar^2 Z^2} \right) - \left(\frac{\alpha(E_1 - E_2)}{\hbar} - \left(\frac{9}{2} \alpha + \frac{\alpha(E_1 - E_2)}{\hbar \varepsilon} \right) \frac{2^{14} e^2 |\mathbf{E}_0|^2 a_0^2}{9^5 \varepsilon \hbar^2 Z^2} \right), \quad (10)$$

$$\beta_2 = -i \frac{2^{14} e^2 |\mathbf{E}_0|^2 a_0^2}{9^5 \varepsilon \hbar^2 Z^2} - \left(\frac{9}{2} \alpha + \frac{\alpha(E_1 - E_2)}{\hbar \varepsilon} \right) \frac{2^{14} e^2 |\mathbf{E}_0|^2 a_0^2}{9^4 \varepsilon \hbar^2 Z^2}, \quad (11)$$

if one assumes that the absolute value of the frequency difference ε is significantly greater than the width of levels and the Stark shift.

In Eqs. (9)–(11) \mathbf{E}_0 is the external field amplitude at the frequency ω ; \mathbf{K} is the wave vector of the light wave; \mathbf{R} is the radius-vector of the atom center of mass.

Taking into account these expressions, one can select the low frequency component accurate to the values of the greater order of smallness:

$$\langle \tilde{\mathbf{d}}(t) \rangle = e a_0 (9 m V_z a_0 / Z^2 \hbar) \cos(\omega_0 t - f_0) \quad (12)$$

of the dipole moment of a hydrogen-like atom

$$\langle \mathbf{d}(t) \rangle = \langle \psi(\mathbf{r}, t) | \mathbf{d} | \psi(\mathbf{r}, t) \rangle / \langle \psi(\mathbf{r}, t) | \psi(\mathbf{r}, t) \rangle, \quad (13)$$

if one assumes the long duration of the action of an external radiation (t is significantly greater than the lifetime of the excited state).

This induced dipole moment is caused by the interference of the quantum states excited by a light wave.

Here the following should be noted. If one solves the Maxwell equation for the light wave of the frequency ω propagating in the medium of the hydrogen-like atoms under consideration, in the approximation of the slowly varying envelope,¹ it is

easy to obtain, within the accepted assumptions on the external field, that this wave coincides with the light wave of the external radiation source.

To derive the expression for the electromagnetic wave of the frequency ω_0 , let us accept some physically obvious assumptions. Let us assume that there is some synchronization of oscillations of the neighbor atoms. The mechanism of such a synchronization has been mentioned above.

In addition, let us take into account the following: when solving Eq. (4), we have separated the external fields to strong and weak ones. This results in the fact that the influence of electromagnetic fields of frequencies different from ω_0 on the dipole moment of the atom practically is not taken into account in the wave function (5). Within the approximation chosen, it is possible to ignore possible transfer of energy from the waves of Raman frequencies to the principal wave. As a result, the following expression is correct for the low-frequency polarization component excited at a given point:

$$\mathbf{P}(t) = N \langle \tilde{\mathbf{d}}(t) \rangle. \quad (14)$$

Here N is the number density of atoms at a given point that have the same oscillation phase.

As known,¹ in the dipole approximation one can write the electric field varying in time at a long distance from the ensemble of atoms in the form:

$$\mathbf{E}(\mathbf{r}_0, t) = \frac{1}{c^2 |\mathbf{r}_0 - \mathbf{r}_1|} \left\{ \left[\frac{\partial^2}{\partial t^2} \mathbf{P} \left(t - \frac{|\mathbf{r}_0 - \mathbf{r}_1|}{c} \right) \times \mathbf{n} \right] \times \mathbf{n} \right\}. \quad (15)$$

Here we introduce the following designations: \mathbf{n} is the unit vector along the direction of observation, \mathbf{r}_0 is the radius-vector of the point of observation, \mathbf{r}_1 is the radius-vector of the point of the dipole moment.

To obtain the total wave disturbance created by a long object at the point of observation, it is necessary to integrate Eq. (15) over the volume occupied by the ensemble of atoms.

Let us accept for certainty that the atom oscillations synchronize due to propagation of some wave in the medium, i.e. let us assume that f_0 changes from point to point according to the law:

$$f_0 = \mathbf{K}_0 \cdot \mathbf{r}_1, \quad (16)$$

where \mathbf{K}_0 is the wave vector of the synchronization wave.

In addition, let us assume that atoms occupy the volume that is a parallelepiped extending from $-l$ to l along the x -axis and from $-b$ to b along the y - and z -axes (the length along the x -axis is significantly greater than the value b). Then one can obtain for the electromagnetic wave generated at the frequency ω_0

$$\begin{aligned} \mathbf{E}_s(\mathbf{r}_0, t) = & [\mathbf{n} \times [\mathbf{k} \times \mathbf{n}]] \frac{8c}{\omega_0 |\mathbf{r}_0|} V_z N \left(\frac{9 e m a_0}{\hbar Z^2} \right) \times \\ & \times \frac{\sin(\omega_0 b n_{0x}/c)}{n_{0x}} \frac{\sin(\omega_0 b n_{0y}/c)}{n_{0y}} \frac{\sin(\omega_0 b n_{0z}/c)}{n_{0z}} \times \\ & \times \cos(\omega_0 t - |\mathbf{K}_0| |\mathbf{r}_0|), \end{aligned} \quad (17)$$

where \mathbf{k} is the unit vector along the z -axis; and n_{0i} are the values determined by the formulas:

$$n_{0x} = \cos \alpha - (c/v_0) \cos \alpha_1; \quad (18)$$

$$n_{0y} = \cos \beta - (c/v_0) \cos \beta_1; \quad (19)$$

$$n_{0z} = \cos \gamma - (c/v_0) \cos \gamma_1, \quad (20)$$

where v_0 is the phase speed of the synchronization wave; $\cos \alpha$, $\cos \beta$ and $\cos \gamma$ are the direction cosines of the vector \mathbf{n} ; and $\cos \alpha_1$, $\cos \beta_1$ and $\cos \gamma_1$ are direction cosines of the vector \mathbf{K}_0 .

It is easy to see, that induced electromagnetic wave of the frequency ω_0 always appears at the presence of the wave synchronization. Its amplitude is mostly determined by the values of the parameters n_{0i} . In the case when these parameters are equal to zero, the amplitude takes its maximum value. This condition can be interpreted as the synchronization condition for the low-frequency electromagnetic wave. However, all three parameters cannot be equal to zero for $v_0 \neq c$. In this case the coefficient n_{0x} is most essential. It follows from this that when an acoustic wave has been generated in the medium containing the hydrogen-like atoms, there are the directions where the IR radiation is maximum in the presence of an external high-frequency radiation.

It is interesting to note that when v_0 is equal to the light speed, the synchronism conditions are satisfied simultaneously for all three parameters. Then the amplitude of the wave of the frequency ω_0 takes the maximum possible value and linearly depends on the volume occupied by atoms.

As a conclusion let us note the following. Formally we have considered an ideal problem. However, it is quite close to the real conditions of the experiments on light scattering. This gives the basis for supposition that the results obtained describe one of the mechanisms of thermal radiation of the objects illuminated by the external radiation source.

If then one takes into account that the excitation of oscillation degrees of freedom of molecules and crystals is possible under the effect of low-frequency radiation, one can consider this mechanism as one of the channels for dissipation of the radiative energy and heating the substance.

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