Low-frequency emission in resonant processes

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Abstract. The paper shows analytically that any resonant interaction of quantum particles in media having no symmetry centre with a high-frequency exciting field produces low-frequency radiation. Media particles may not necessarily have a constant dipole moment. Conditions are identified when carrier frequency of low-frequency radiation coincides with nutational oscillations of resonant level populations and is readily controlled both by high-frequency resonant radiation intensity and by resonance detuning.

1. Introduction
In recent years generation of low-frequency radiation whose carrier frequencies are in the terahertz range has been actively explored with the purpose of practical application. Among the nonlinear-optical resonant phenomena, research works [1-5] studied resonant processes of interaction of optical (and other “high-frequency”) waves with particles having permanent dipole moment. Research is focused not only on the generation of terahertz radiation, but also on the effect of permanent dipole moment on the excitation pulse propagation [6-9].

The paper has demonstrated analytically that for generation of low-frequency radiation in resonant-optical processes, including terahertz radiation, it is not necessary to study environments whose particles have permanent dipole moment. Generation of low-frequency radiation is also possible in ordinary environments in conditions of simple one-photon resonant interaction between exciting coherent pulses and an environment. It is necessary that resonant particles have non-resonant energy levels of indefinite parity, which may be formed, e.g., by levels with different parity but similar energy (which is characteristic of the media with quadratic nonlinear response). Meanwhile, in some certain conditions carrier frequency of low-frequency radiation precisely equals frequency of nutational oscillations of resonant level populations of quantum particles. Since frequency of nutational oscillations is proportional to the square root of resonance pumping intensity, it provides for a simple method of controlling carrier frequency of low-frequency radiation up to terahertz radiation generation.

As far as the intensity of generated low-frequency radiation is concerned, it proves to be as low as intensity of any two-quantum process in an electromagnetic wave.

The obtained results followed from considerations of resonant interactions up to the second-order perturbation theory. Second order of smallness is certain to be disadvantageous compared to the case of low-frequency generation for environments having particles with permanent dipole moment. At the same time, theoretical consideration of generation processes in environments containing resonant and...
populated levels with no permanent dipole moment is more correct since there arise no questions concerning phase transition in condensed environments whose particles have permanent dipole moments and which are not taken into account by low-frequency generation investigations. Both the intensity smallness of the explored radiation and the coincidence of its propagation direction with an exciting pulse direction is likely to hinder experimental detecting this radiation against powerful pumping.

Studying resonant interaction of coherent radiation with matter in theory and experiment, the so-called optical nutation [10,11] is referred to as one of the fundamental effects. Nutational oscillations are meant as oscillations of radiation intensity, having passed through optically thin resonant environment near its front edge if the exciting pulse envelope has a squared shape. At the same time, radiation intensity is considered to be averaged over a period of fast oscillations of an electromagnetic field. Frequency of nutational oscillations, multiplied by Planck constant, represents interaction energy of electromagnetic radiation with a resonant particle. Therefore, a coherent field with a squared shape envelope passing through the optically thin resonance environment has two frequency characteristics: radiation carrier frequency and quantum reradiation frequency within the field of this radiation (nutation), with radiation carrier frequency being far higher than that of nutational oscillations.

The paper has shown how radiation of carrier frequency, being equal to frequency of nutational oscillations, is formed under certain conditions in environments with no permanent dipole moment. Generally, it is not surprising because the paper defines that in a resonant wave field the particle dipole moment contains intermodulation components. A consistent theory of resonant interaction of coherent radiation with the particle environment with account of non-resonant particle states has been introduced; common expressions for effective operators of dipole moment of environmental particles have been derived. Application of the developed theory is a new possibility of controlling carrier frequency of the generated radiation by means of intensity change of an exciting wave. In particular, the considered process can be useful for generating quasi-monochromatic terahertz (or subterahertz) radiation.

2. Algebraic perturbation theory
Let propagate a coherent wave towards the axis $Z$ whose intensity $E$ of electric field is represented as a slowly changing amplitude $\varepsilon$ and fast changing phase defined by a wave vector $\mathbf{k} \parallel Z$ and carrier frequency $\omega$:

$$E = \varepsilon \exp[i(kz - \omega t)] + c.c. \equiv \varepsilon \exp[-i\Phi] + c.c.$$ Polarization effects will be neglected assuming that electric vector is parallel to axis $X$. Letters $c.c.$ designate terms complexly conjugated with the previous term. $\Phi = \omega t - kz$ is the phase of a coherent wave.

Semispace $Z \geq 0$ is filled with environment consisting of resonant particles. Particle resonance levels consist of ground state $|g\rangle$ and excited state $|e\rangle$. Frequency $\omega_{eg}$ of transition $|e\rangle \rightarrow |g\rangle$ is close to frequency $\omega$. $|\omega_{eg} - \omega| < \omega$. Generally, in analogous problems transition $|e\rangle \rightarrow |g\rangle$ is referred to as optically allowed transition supposing, firstly, that dipole transition moment $d_{eg} \neq 0$, and, secondly, that states $|e\rangle$ and $|g\rangle$ are characterized by different parity. These suppositions may also be accepted in our consideration, however, requirement for different parity may be omitted – it is enough that only $d_{eg} \neq 0$ exists, but permanent dipole moment is absent $d_{ee} = d_{gg} = 0$.

Neglecting interaction between particles, dipole moment of environment unit volume $P$ can be expressed as

$$P = nSppd,$$
where \( n \) is the density of resonance particles per unit volume, \( d = \sum_{kj} d_{kj} \) is the operator of dipole moment of resonance particle, \( |E_k> < E_j| \) is the quantum state of resonance particle, which may be degenerated, and \( \rho \) is its density matrix. Polarization of environment \( \rho \) serves the effective Hamiltonian \( H \) and the operator \( c |E_k> < E_j| \) is the quantum state of resonance particle, which may enter Maxwell equation

\[
(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2})E = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\rho.
\]

In ignoring relaxation and spontaneous particle radiation as well as with account of electrodipole approximation in interaction of particle with a coherent field, equation for density matrix takes a standard form

\[
ih\frac{\partial \hat{\rho}}{\partial t} = [H_0 - Ed, \rho].
\]

Following the general method [10], let us transform density matrix:

\[
\hat{\rho} = U\rho U^+, \quad U = e^{-is}, \quad S^+ = S.
\]

Transformed density matrix will satisfy kinetic equation

\[
ih\frac{\partial \hat{\rho}}{\partial t} = [\hat{H}, \hat{\rho}]
\]

with the transformed Hamiltonian

\[
\hat{H} = U(H_0 - Ed)U^+ - ihU\frac{\partial}{\partial t}U^+.
\]

Such representation is convenient for developing perturbation theory. Let us expand \( \hat{H} \) and \( S \) into a series according to the interaction with a coherent field, marking with a superscript a size of order of terms with regard to coupling constant of the coherent field:

\[
S = S^{(1)} + S^{(2)} + \ldots, \quad \hat{H} = \hat{H}^{(0)} + \hat{H}^{(1)} + \hat{H}^{(2)} + \ldots
\]

Taking the Baker–Hausdorff formula [11] into account, we obtain

\[
\hat{H}^{(0)} = H_0, \quad \hat{H}^{(1)} = -Ed - i[S^{(1)}, \hat{H}^{(0)}] + h\hat{c}S^{(1)} / \hat{c}t, \quad \hat{H}^{(2)} = \frac{i}{2}[S^{(1)}, Ed] - \frac{i}{2}[S^{(1)}, \hat{H}^{(1)}] - i[S^{(2)}, \hat{H}^{(0)}] + h\hat{c}S^{(2)} / \hat{c}t.
\]

The main idea to find terms \( S^{(i)} \) and the effective Hamiltonian \( H^{eff} = \hat{H}^{(0)} + \hat{H}^{(1)} + \hat{H}^{(2)} \) serves the absence of fast-changing in time terms in \( \hat{H}^{(i)} \) in interaction representation. This leads to “correctly” changing in time terms in the following expressions:

\[
\hat{H}^{(1)} = -\varepsilon \exp(\zeta \Phi) d_{g}\varepsilon |E_g > < E_g| + H.c., \quad S^{(1)} = \sum_{kj} d_{kj} \left( \frac{\varepsilon \exp(-i\Phi)}{\omega_{jk} - \omega} + \frac{\varepsilon' \exp(i\Phi)}{\omega_{jk} + \omega} \right) |E_j > < E_k|,
\]

\[
\hat{H}^{(2)} = -|\varepsilon' | \sum_{k} \Pi_{jk} (\omega) |E_k > < E_k|,
\]

where main parameters of the theory of optical resonant processes [11] are involved.
\[ \Pi_{ij}(\omega) = \sum_{j} \frac{d_{kj}d_{nj}}{\hbar} \left( \frac{1}{\omega_{nk} + \omega} + \frac{1}{\omega_{nj} - \omega} \right) = \Pi_{jk}^*(-\omega). \]

Letters \( H.c \) designate terms which are Hermitean-conjugated to the previous terms. Stroke of the summation symbol indicates that resonant terms \( \omega_{nk} \approx \omega \approx 0 \) are excluded in summation.

It is important to stress that \( \Pi_{eg}(\omega) \neq 0 \) for \( |e > \) and \( |g > \) states, even in the case of different parity of resonant states \( |e > \) and \( |g > \), since in summation in expression for \( \Pi_{eg}(\omega) \) all states are accounted, including degenerate states and states with no certain parity. Moreover, the theory does account for both certain parity and absence of parity of resonant states.

Polarization of environment, expressed in terms of the transformed density matrix \( P = nSp\tilde{\rho}d \), is defined by the effective operator of dipole moment

\[ \tilde{d} = e^{-i\pi}de^{i\phi} = d - i[S,d] - \frac{1}{2}[S,[S,d]] + \ldots = \sum_{kj} \tilde{d}_{kj} |E_k > < E_j|, \]

\[ \tilde{d}_{kj} \approx d_{kj} + \varepsilon \Pi_{kj}(\omega)e^{-\varepsilon\phi} + \varepsilon^* \Pi_{kj}^*(-\omega)e^{\varepsilon\phi}. \] (1)

Characteristic feature of polarization of resonant environment with \( \Pi_{eg}(\omega) \neq 0 \), but in absence of permanent dipole moment \( d_{ee} = d_{gg} = 0 \), is the presence of several characteristic frequencies in the field of resonant pulse \( (\tilde{p}_{eg} = R_{eg}e^{-\varepsilon\phi}, R_{eg} \text{ is the slowly changing in time amplitude of coherence of a resonant particle})

\[ P = n\tilde{p}_{eg}\tilde{d}_{ge} + H.c. = nR_{eg}d_{ge}e^{-\varepsilon\phi} + n\varepsilon\Pi_{ge}(\omega)e^{-2\varepsilon\phi} + n\varepsilon^*R_{eg} \Pi_{ge}^*(-\omega) + \varepsilon R_{ee} \Pi_{ee}(\omega)e^{-\varepsilon\phi} + \varepsilon^* R_{eg} \Pi_{ge}(\omega)e^{\varepsilon\phi} + H.c. \] (2)

Among them the term of interest, which takes the following form

\[ P_{\lambda} = n\varepsilon^*R_{eg} \Pi_{ge}^*(-\omega) + H.c. \]

is characterized only by frequencies of slowly changing coherence amplitude \( R_{eg} \).

3. **Generation of low-frequency radiation in proximity of front edge of resonant pulse**

Let the coherent pulse entering the resonant environment be rectangular in shape \( (a = a^*) \):

\[ \varepsilon (z = 0, t) = \begin{cases} a, & t \geq 0, \\ 0, & t < 0. \end{cases} \]

Then for optically thin environments of length \( L \), when the feedback effect on the passing pulse can be neglected, amplitude of coherence of resonant particles with resonance detuning \( \Delta \) takes the form

\[ R_{eg} = \frac{ad_{eg} / \hbar}{\Omega} \left[ \frac{\Delta}{\Omega}(1 - \cos \Omega t) + i \sin \Omega t \right], \]

\( \Omega = \sqrt{\Delta^2 + \Lambda^2}, \ \Lambda = 2 |ad_{eg} / \hbar| \). Let us suppose that distribution of resonant particles throughout detunings from rigid resonance is given by some function \( F(\Delta) \).

The considered amplitude \( R_{eg} \) of coherence of resonant atoms provides for oscillations of environment polarization whose central frequency coincides with frequency \( \Lambda \). This is exactly the frequency of nutational oscillations of resonant level populations.
\[ P_\lambda = n \Pi_{ge}(-\omega) \frac{a^2 d_{ge}}{\hbar} < \frac{1}{\Omega} \left( \frac{\Lambda}{\Omega} (1 - \cos \Omega \tau) + i \sin \Omega \tau \right) > + H.c. \] (3)

Angular brackets indicate averaging over spread of detuning with a distribution density \( F(\Delta) \).

Thus, periodic dynamics of quantum populations over Rabi frequency gives rise (by means of a term in an effective dipole moment of quantum particle) to polarization oscillations over nutation frequency. In order to find intensity of electric field \( E_\lambda \), emitted by these polarization oscillations, it is necessary to solve the Maxwell equation.

Radiation over frequency of nutational oscillations of resonant levels is derived from approximation of unidirectional waves [12]. Then the Maxwell equation is reduced to [12]:

\[ \left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) E_\lambda = -\frac{2\pi}{c} \frac{\partial}{\partial t} P_\lambda. \]

For optically thin environments of length \( L \) we obtain that beyond the limits of slowly changing electric field amplitude, generated at low frequency, takes the form:

\[ E_\lambda = -\frac{\pi n L h c^{-1}}{2} \Pi_{ge}(-\omega) \frac{d_{ge}}{\Delta^2} \frac{\Delta}{\Omega} \left( \frac{\Lambda}{\Omega} \sin \Omega (t - \frac{z}{c}) + i \cos \Omega (t - \frac{z}{c}) \right) + c.c. \] (4)

Broadly speaking, parameters of resonant environment entering coefficient \( \Pi_{ge}(-\omega)/d_{ge} \), are not purely real values. So

\[ \Pi_{ge}(-\omega)/d_{ge} = \text{Re}(\Pi_{ge}(-\omega)/d_{ge}) + i \text{Im}(\Pi_{ge}(-\omega)/d_{ge}). \]

It follows from formula (4) that central frequency of low-frequency oscillations coincides with frequency of nutational oscillations of resonant level populations.

At resonance detuning with no energy level spread (or at narrow frequency spread compared with \( \Lambda \)) and \( \text{Im}(\Pi_{ge}(-\omega)/d_{ge}) = 0 \):

\[ E_\lambda = -\pi n L h c^{-1} \Pi_{ge}(-\omega) \frac{d_{ge}}{\Delta^2} \frac{\Delta}{\Omega} \sin \Omega (t - \frac{z}{c}). \]

Frequency of low-frequency oscillations coincides with frequency of nutational oscillations.

Frequency spread of resonant particles leads to attenuation of low-frequency radiation despite the fact that its exciting radiation continues affecting resonance environment. In addition, in the simplest case of exact resonance \( \Delta = 0 \) and the pure real meaning of value \( \Pi_{ge}(-\omega)/d_{ge} \) the low-frequency radiation is absent \( E_\lambda = 0 \). This fact, along with coincidence of direction of studied low-frequency radiation and direction of pumping resonant wave, hinders experimental observation of low-frequency radiation (4).

Practical application of studied low-frequency radiation is the possibility of tuning of radiation carrier frequency by means of change in pumping intensity. For frequency of low-frequency radiation under discussion to be in terahertz region \( \Lambda \sim 10^{12} \) s\(^{-1} \) (what is required in many problems) exciting resonant fields of order \( I \sim 10^8 \) Wt/cm\(^2\), are needed if one estimates that \( d_{ge} \sim 10^{-18} \) CGSE and takes into account the following expression \( \Lambda = (8\pi L c^{-1} h^{-2})^{1/2} d_{ge} \).

4. Conclusion

The result under discussion has so far not received due attention in theoretical research works on nonlinear-optical events in resonant environments. Obviously, it is connected with the fact that processes of environment resonance interaction are treated by researchers in terms of two-level models
constructed not on the basis of the strict perturbation theory but merely by suppression of non-resonant levels [13,14]. To interpret two-level models physicists generally use averaging methods while considering terms that change fast and slowly in time (such as Bogoliubov-Mitropolsky method [15,16]). But in case your attention is focused only on resonant effects in the averaging method it is hard to notice, due to its awkwardness, other parametric processes generated by resonant interaction.

The averaging method provides for correct effective equations. However, in case of optical problems it is difficult to present the whole picture of parametric interactions of waves in a resonant environment in a simple and vivid way, using the above indicated method. In this paper a more simple and effective approach is based on algebraic perturbation theory [17,18], which was developed in works [11,19] as applied to nonlinear-optical problems.

Consistently applied, the theory of resonant interaction in terms of algebraic perturbation theory [11] allows not only explain rigidly the approximation of two-level models but also in a simpler way, compared to averaging methods, to find expressions for theory parameters which possess characteristics of all non-resonant levels of particles in resonant environment along with characteristics of all resonant levels. In addition, the method allows embracing the variety of parametric interactions in resonant environment and identify the described effect of low-frequency radiation at the frequency of nutational oscillations of resonant level populations of environment.

In conclusion, it should be noted that the phase switching method applied to the exciting high-frequency field [20] can be used to provide for an effective resonant interaction of the medium with an exciting field of a rectangular shape.

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