Delay-dependent amplification of a probe pulse via stimulated Rayleigh scattering

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Stimulated Rayleigh scattering of pump and probe light pulses of close carrier frequencies is considered. A nonzero time delay between the two pulses is shown to give rise to amplification of the delayed (probe) pulse accompanied by attenuation of the pump, both on resonance and off resonance. In either case, phase-matching effects are shown to provide a sufficiently large gain, which can exceed significantly direct one-photon-absorption losses.

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Rayleigh scattering in continuous media is attributed to scattering off “nonpropagating modulations of material observables” [1]. The elementary quantum-mechanical process underlying Rayleigh scattering is the two-photon emission-absorption transition of an atom such that the initial and final atomic states coincide [2]. In the approximation where both atomic recoil and motion of the atoms are ignored, the frequencies of the emitted and absorbed photons are equal though the directions of their wave vectors can be different. The concept of stimulated Rayleigh scattering arises when one considers the same atomic transitions but in the presence of two specified waves, pump and probe, such that stimulated emission of either pump or probe photons prevails over spontaneous emission.

In a traditional formulation of the problem of stimulated Rayleigh scattering [2], both pump and probe wave are assumed to be monochromatic plane waves. In this case, for an ensemble of stationary recoilless atoms, there is no net Rayleigh scattering: the probability of the direct process (absorption of a pump photon and emission of a probe photon) is exactly equal to that of the inverse process (emission of a pump photon and absorption of a probe photon). Hence, there is no net conversion of the two species of photons into each other. Here, we will consider a different situation: we will assume that pump and probe fields are finite pulses, whose centers are separated by the variable delay $\Delta t$. It will be shown that in this case a photon of the pump wave can be transformed into a photon of the (delayed) probe wave. The efficiency of this process can be very high, owing to phase matching effects of coherent stimulated Rayleigh scattering off an ensemble of atoms.

In the case we consider, the phases of pump and probe waves are completely uncorrelated. Owing to this assumption, only the probabilities of the direct and inverse processes described above are meaningful quantities, rather than a coherent sum of transition amplitudes. The complete lack of coherence between the pump and the probe wave distinguishes this case from pulse propagation in a resonant medium [3,4] (otherwise, the pump and the probe could be combined into one pulse whose propagation one could study). Also, evidently, stimulated Rayleigh scattering is conceptually different from bichromatic pulse propagation in three-level media, where the elementary underlying processes are $\Lambda$ or Raman transitions between different initial and final atomic states [5,6].

It should also be mentioned that in our case the pump-to-probe transformation of photons or, in other words, the amplification of the probe wave is not connected with any kind of population inversion: initial- and final-state populations are equal because initial and final states are identical. In this sense, stimulated Rayleigh scattering is reminiscent of the well-known inversionless lasers [1], even though the physics of amplification in our scheme and in inversionless lasers are completely different.

A closely related problem was investigated in a recent work on amplification of high-order harmonics of a strong laser field [8]. However, the latter is a rather complicated process, whose theoretical description is based on various assumptions and approximations, which shadow the physics of the phenomenon. On the other hand, the prob-
lem formulated above is so fundamental that it deserves description of its simplest manifestation, which occurs in Rayleigh scattering to be considered in this letter.

So, let the electric-field strength of the pump and the probe pulse be given by
\[
\begin{align*}
\varepsilon(t) &= \varepsilon_0(t) \cos(\omega_0 t - \mathbf{k} \cdot \mathbf{r}), \\
\tilde{\varepsilon}(t) &= \tilde{\varepsilon}_0(t) \cos(\tilde{\omega}_0 t - \tilde{\mathbf{k}} \cdot \mathbf{r}),
\end{align*}
\]
respectively, where \(|\mathbf{k}| = \omega_0/c\) and \(|\tilde{\mathbf{k}}| = \tilde{\omega}_0/c\). The fields of both pulses are assumed to be linearly polarized along the same direction.

By assuming that an atom, located at \(\mathbf{r} = \mathbf{R}_j\), is initially in its ground state and the fields \(\varepsilon\) and \(\tilde{\varepsilon}\) are weak enough, we calculate the second-order perturbation-theory amplitude of the transitions involved in stimulated Rayleigh scattering and present it in the form
\[
A^j = A^j_{\text{em}} + A^j_{\text{abs}},
\]
where \(A^j_{\text{em}}\) and \(A^j_{\text{abs}}\) are, respectively, the transition amplitudes involving stimulated emission and absorption of a probe-wave photon \(E\).

Identification of the two terms in the sum \(A\) as the amplitudes of stimulated emission and absorption is related to the sign in front of the carrier frequency \(\tilde{\omega}_0\) of the probe in the exponent on the right-hand side of Eq. (4). This separation of emission and absorption is illustrated by the four diagrams of Fig. 1. The first two diagrams of Fig. 1 \([a]\) and \([b]\) correspond to emission of a probe-wave photon with frequency \(\tilde{\omega}\) (the arrows of the lines pointing down), whereas the other two \([c]\) and \([d]\) correspond to absorption (the arrows pointing up). The two diagrams in each line of Fig. 1 correspond to the two terms in big parentheses on the right-hand side of Eq. (4). Only the near-resonant terms \([b]\) and \([c]\) make a significant contribution. These two terms are proportional to \(\tilde{\varepsilon}_0(t)\varepsilon_0(t')\) and \(\varepsilon_0(t)\tilde{\varepsilon}_0(t')\), respectively. Since in Eq. (4) we always have \(t > t'\), already this very general expression shows that stimulated emission can prevail over absorption provided the probe pulse is retarded with respect to the pump pulse, because in this case \(\varepsilon_0(t)\varepsilon_0(t') > \tilde{\varepsilon}_0(t)\tilde{\varepsilon}_0(t')\) if pulse durations are short enough.

By expanding \(\varepsilon(t)\) and \(\tilde{\varepsilon}(t)\) in terms of Fourier integrals
\[
\left\{ \varepsilon(t), \tilde{\varepsilon}(t) \right\} = \int_{-\infty}^{\infty} d\omega \exp(i\omega t) \left\{ \varepsilon_\omega, \tilde{\varepsilon}_\omega \right\},
\]
we can reduce Eq. (4) to the much simpler form
\[
A_{\text{em/abs}} = 2\pi i \int_0^\infty d\omega \alpha(\omega) \left\{ \varepsilon_\omega, \tilde{\varepsilon}_\omega \right\},
\]
where \(\alpha(\omega)\) is the complex atomic polarizability
\[
\alpha(\omega) \equiv \alpha_1(\omega) + i \alpha_2(\omega) = \sum_n |d_{0n}|^2 \times \left( \frac{1}{E_n - E_0 - \omega - i\delta} + \frac{1}{E_n - E_0 + \omega - i\delta} \right) \downarrow_{\delta \rightarrow 0}.
\]

For a gas of atoms two-photon absorption-emission transitions in different atoms correspond to the same initial and final state but different intermediate states of the total multimode system. Summation over intermediate states includes summation of the probability amplitudes \(A^j_{\text{em/abs}}\) of the various atoms. The total probability of emitting a probe photon \(\tilde{\omega}\) is given by the difference of the probabilities of its stimulated emission and absorption summed over all atoms,
\[
u_T^\text{tot} = \left| \sum_j A^j_{\text{em}} \right|^2 - \left| \sum_j A^j_{\text{abs}} \right|^2 \equiv \nu_T F,
\]
where \(F\) is the phase-matching factor.

and \(w_T\) the single-atom total emission probability, \(w_T = |A_{\text{em}}|^2 - |A_{\text{abs}}|^2\).

If \(w_T\) and \(\nu_T^\text{tot}\) are positive, the gain \(G\) of the probe wave in an atomic gas is determined as the ratio of the average total energy \(h\omega_0\nu_T^\text{tot}\) gained by the probe wave over its incident energy \(V\varepsilon_0^2/8\pi\),
\[
G = \frac{8\pi \hbar \omega_0}{V} \nu_T^\text{tot} = \frac{8\pi \hbar \omega_0}{\varepsilon_0^2} n_a \frac{F}{N_a} w_T,
\]
where \(V\) is the interaction volume, \(N_a\) the total number of atoms and \(n_a = N_a/V\) their density.

The phase-matching factor \(F\) arises from coherent (in-phase) emission of photons by different atoms, and it can be rather large. For \(\mathbf{k} = \mathbf{k}_0\), it assumes its maximal value \(F_\text{max} = N_a^2\). However, in order that the pump and the probe mode can be experimentally distinguished, we will
consider $k \approx \tilde{k}$. If the length of the interaction region in the direction of $k - \tilde{k}$ is $\leq 1/|k - \tilde{k}|$, we still have $F \approx F_{\text{max}}$. In that case, the coherent gain $\tilde{g}$ differs from the incoherent one by the factor $F/N_0 \gg 1$.

Let us now specialize the pulse envelopes $\varepsilon_0(t)$ and $\varepsilon_0(t)$ to the Gaussians

$$
\varepsilon_0(t) = \varepsilon_0 \exp\left(-\frac{t^2}{2\tau^2}\right), \quad \tilde{\varepsilon}_0(t) = \varepsilon_0 \exp\left(-\frac{(t - \Delta t)^2}{2\tau^2}\right),
$$

where $\Delta t$ is the delay of the probe pulse. For simplicity, we assume that the pump and the probe pulse have identical durations and carrier frequencies, $\tilde{\omega} = \omega \equiv \omega_0$ and $\tau = \tau$. Then, the probability amplitudes (11) become

$$
A_{\text{abs}}(\omega) = i \frac{\varepsilon_0 \tilde{\varepsilon}_0 \tau^2}{4} \int_0^\infty d\omega \alpha(\omega) \times \exp\left[-(\omega - \omega_0)^2/\tau^2 \mp i(\omega - \omega_0)\Delta t\right].
$$

First, we assume that the carrier frequencies of both pulses are resonant with some bound-bound atomic transition, $\tilde{\omega}_0 = \omega_0 = E_1 - E_0$. In this case, the polarizability (1) can be approximated by its resonant part

$$
\alpha(\omega) \approx -\frac{|d_{10}|^2}{(\omega - \omega_0) + \frac{i\delta_0}{2}},
$$

where $\Gamma$ is the width of the level $E_1$, which we put in by hand (a calculation to all orders in the pump would furnish it automatically). Under these conditions, directly from Eqs. (12) and (13) we get

$$
w_T = (\tau \Omega R)^2 (\tau \tilde{\Omega} R)^2 J,
$$

where $\Omega R = \frac{1}{2} \varepsilon_0 |d_{01}|$ and $\tilde{\Omega} R = \frac{1}{2} \tilde{\varepsilon}_0 |d_{01}|$ are the pump- and probe-wave Rabi frequencies, and $J$ is a dimensionless function of the dimensionless variables $t_0 = \Delta t/\tau$ and $r = \tilde{\omega}/\Omega R$,

$$
J = -\frac{1}{\Gamma} \frac{d}{dt_0} \left[ \int_{-\infty}^{\infty} \exp(-x^2 + it_0 x) \frac{x^2 + \gamma^2}{4} \right]^2.
$$

For $\gamma \gg 1$, Eqs. (13)–(15) yield

$$
w_T = \frac{16 \pi \Omega R^2 \tilde{\Omega} R}{\Gamma^3} \Delta t \exp \left\{ -\frac{1}{2} \left( \frac{\Delta t}{\tau} \right)^2 \right\}.
$$

Increasing the width $\Gamma$ results in a rather quick decrease of the two-photon Rayleigh-scattering probability $w_T$. This restricts (for $\Gamma / \tau > 1$) the region of time delays $\Delta t$ where $w_T$ is not small to the range $|\Delta t| \lesssim 1/\tau$ (cf. Fig. 2).

The antisymmetric shape (with respect to $\Delta t$) of the gain (10) is reminiscent of the free-electron laser (FEL), whose gain is antisymmetric with respect to the detuning $\delta\omega = \omega - \Delta \omega$. There are other interesting relations between our scheme and the FEL that will be discussed elsewhere.

It should be noted that the width $\Gamma$ can be significantly larger than a typical single-atom radiative width $\Gamma_{\text{r}} \sim 10^8 \text{s}^{-1}$, owing to coherent spontaneous forward emission by coherently excited atoms. Actually, in addition to the two-photon transition we here consider, the pump field also provides a real population of the resonant level $E_1$. Atoms excited in such a way can emit photons spontaneously. This is a sequential two-step absorption and emission process, which is different from the coherent quantum-mechanical two-photon transition considered above. But, if the phases of the atomic excitation do not change significantly during the duration of the pulse, spontaneous emission is characterized by the same phase-matching factor (13) as stimulated emission. For the former, however, the wave vector $k$ can have arbitrary direction. For this reason, the phase-matching factor for spontaneous emission appears to be given by $F_{\text{sp}} = \int F(n) dN_n$, where $n = k / k$. In other words, laser excitation prepares a coherent Dicke-type ensemble of excited atoms, whose spontaneous emission can be strongly enhanced in comparison with the case of incoherently excited atoms (10). For a laser focus with waist $d$ and length $L \sim d^2 / \lambda$, where $\lambda = 2\pi / k$ is the wavelength, a simple estimate based on replacing the summation in Eq. (13) by an integration gives $F_{\text{sp}} \sim (d / L)^2 N^2_a$. The product $\Gamma F_{\text{sp}}$ determines the total number of photons emitted per unit time by the ensemble of atoms in the focal volume. Divided by $N_a$ it gives the rate of transitions per single atom or a phase-matching-modified radiative width of the level $E_1$: $\Gamma = \Gamma_{\text{r}} F_{\text{sp}} / N_a \sim \Gamma_{\text{r}} (d / L)^2 N^2_a$. For example, for $\lambda \sim 10^{-4}$ cm, $d \sim 10^{-3}$ cm, $L \sim 10^{-2}$ cm, and $N_a \sim 10^{16}$ cm$^{-3}$ we get $N_a \sim 10^8$, $F_{\text{sp}} \sim 10^{14}$ and $\Gamma \sim 10^{14}$ s$^{-1}$. Therefore, even for 100-fs pump pulses the parameter $\Gamma / \tau \approx 10 \gg 1$, and the approximation (10) is relevant. For these parameters and for $\Delta t \sim \tau \sim 10^{-13}$ s, $\Omega R \tau \sim \tilde{\Omega} R \tau \sim 10^{-2}$, the gain (16) of the probe wave is of the order of one, $\Gamma \sim 1$. Finally, as the direct one-photon absorption of probe photons does not experience any phase-matching enhancement, it can be checked easily to be much less efficient than the here described two-photon stimulated Rayleigh scattering process.

Next, we will consider the case when the pump- and probe-wave frequencies $\omega$ and $\tilde{\omega}$ exceed the ionization energy $E_b$. For direct numerical calculations with the help of Eqs. (8) and (12) we use the $\delta$-potential model, for which the atomic polarizability is given by (13) $\alpha(\omega) = a(x) / E_b^2$, where $x = \omega / E_b$ and

$$
a(x) = \frac{1}{x^2} + \frac{8}{3x^4} - \frac{4}{3} \frac{(x + 1)^3/2 - i(x - 1)^{3/2}}{x^4}.
$$

In the case $\omega_0 - E_b \gg 1/\tau$ the integral over $\omega$ in Eq. (12) can be calculated analytically to give
\[ w_T = \frac{\pi \varepsilon_0^2 \varepsilon_0^2}{8} \Delta t \exp \left( -\frac{\Delta t^2}{2\tau^2} \right) \times \left( \alpha_1(\omega_0)\alpha_2(\omega_0) - \alpha_2(\omega_2)\alpha_1'(\omega_0) \right). \] (18)

The dependence of \( w_T \) on the delay \( \Delta t \) is similar to that occurring in the resonance case when the width of the resonant level is large (cf. Fig. 2): \( w_T(\Delta t) \) is an odd function and is localized in the range \( |\Delta t| < \sim 1/\tau \).

The dependence of \( w_T \) [Eq. (18)] on \( \omega_0 \) is shown in Fig. 3, and this result shows that the effect is maximal approximately at \( \omega_0 \approx 1.2 E_0 \).

In the case of nonresonant transitions, the gain (11) can easily become as large as in the resonant case. For the same values of \( \lambda, d, \) and \( L \) as in the previous estimates, the value \( G \sim 1 \) is reached at \( n_a \sim 10^{17} \text{ cm}^{-3} \). Again, as the direct losses due to ionization do not experience phase-matching enhancement, they are insignificant on the scale of the energy transfer that is achievable in two-photon stimulated Rayleigh scattering.

In conclusion, stimulated Rayleigh scattering, both resonant with an atomic bound-bound transition or via the continuum, may furnish substantial gain in a pump-probe experiment where the probe pulse is delayed with respect to the pump. This scenario appears to have escaped attention thus far.

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Fig. 1. Diagrams of two-photon transitions involved in stimulated Rayleigh scattering. The ground state with energy \( E_0 \) (solid) and a near-resonant excited state with energy \( E_n \) (dashed) are indicated by horizontal lines. The latter may as well represent the continuum threshold.

Fig. 2. The function \( J(\Delta t/\tau) \) [Eq. (15)], which is proportional to the total probability (14) of resonant stimulated Rayleigh scattering for \( \Gamma \tau = 10 \).

Fig. 3. The function \( \text{Re} a(x) \text{Im} a'(x) - \text{Im} a(x) \text{Re} a'(x) \), which characterizes the dependence of \( w_T(\omega) \) [Eq. (18)].
Normalized delay, $t_0 = \Delta t/\tau$
\[ f(x) = \alpha_1(x)(x) - \alpha_2(x)(x) - \alpha_1'(x) \]

Normalized frequency, \( x = \frac{\omega}{E_b} \)