Anomalous Stimulated Brillouin Scattering via Ultraslow Light

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We study stimulated Brillouin scattering (SBS) in an ultradispersive coherent medium, and show that the properties of SBS change drastically when the group velocity of light in the material approaches or becomes less than the speed of sound. In particular, forward SBS not allowed in a dispersionless bulk medium takes place in the coherent medium.

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It is textbook wisdom that forward stimulated Brillouin scattering (SBS) is forbidden by energy and momentum conservation [1], with the strongest scattering typically in the backward direction. Surprisingly, we find that in ultradispersive coherent media, just the opposite can happen. Namely, maximum scattering is in the forward direction while back scattering is forbidden.

Atomic coherence plays an important role in quantum optics. Coherent resonant nonlinear optics provides new materials that have large optical nonlinearities and small absorption [2]. In particular, recent progress in coherent resonant nonlinear optics has been made possible by electromagnetically induced transparency (EIT) [3], which is the basis for the present work.

The EIT observed in multilevel atomic systems is accompanied by a steep linear dispersion which results in substantial reduction of the group velocity of light to values comparable with (or even less than) the speed of sound in the media. Such slow group velocity ("slow light") has been demonstrated experimentally in cold [4] and hot [5] atomic gases as well as in crystals doped by rare-earth ions [6].

The reduction of resonant absorption together with the steep dispersion opens the possibility for new kinds of resonant interactions between electromagnetic and acoustic waves. For example, it has been shown in [7] that the longitudinal gradient force, acting on a two-level test atom, can be enhanced via spatial compression of an optical pulse moving with ultraslow group velocity in a coherent medium. This enhanced force yields a ballistic atom motion and atom surfing, and a new kind of local ponderomotive light scattering.

On the other hand, as has been shown in [8] taking advantage of the large linear dispersion associated with EIT, it is possible to achieve phase matching between electromagnetic waves and acoustic waves in a dielectric fiber doped by three-level Λ-type ions. This can lead to an increase of efficiency of ponderomotive nonlinear interaction between the electromagnetic waves and holds promise for applications, e.g., production of squeezed light.

The main goal of the present work is to study the properties of light scattering in a coherent medium itself without doping it or using any nonlinearities of a host background material. We here show that, in a medium consisting of three-level Λ-type atoms (Fig. 1), forward SBS is possible. Because of the steep linear dispersion associated with EIT phase matching conditions between the acoustic and electromagnetic waves change dramatically compared with those of media with small dispersion. This allows one to obtain a strong acousto-optical interaction yielding SBS even in the forward direction. This should be observable in atomic gases and in solids doped with rare-earth ions and might increase the sensitivity of existing methods of measurements based on SBS.

In order to set the stage for the present ideas, let us first recall the physics of ordinary SBS. The original work of Brillouin [9], devoted to the study of the scattering of light by thermally excited acoustic waves, as well as subsequent experimental [10] and theoretical [11] investigations of stimulated light scattering by acoustic waves, has shown that the most efficient scattering is achieved in the backward direction. Furthermore, the scattering in the forward direction is not possible; see Eqs. (5) and (6)).

Consider propagation of two plane monochromatic waves $E_{p1}e^{-i(\nu_1 t - \vec{k}_1 \vec{r})}$ and $E_{p2}e^{-i(\nu_2 t - \vec{k}_2 \vec{r})}$ in a dielectric medium. The resonant interaction of this traveling intensity wave with sound vibrations of the medium gives rise

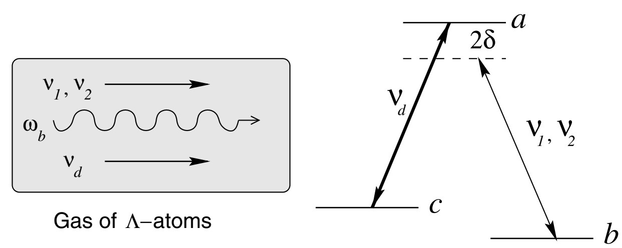

FIG. 1. The scheme of acousto-optical interaction. Probe electromagnetic waves E_{p1} and E_{p2} with carrier frequencies ν_1 and ν_2 ($\delta \gg |\nu_1 - \nu_2|$) and drive wave E_d with carrier frequency ν_d propagate in a coherent medium consisting of three level atoms in Λ configuration. An appropriate choice of the drive field intensity allows one to establish strong interaction between the probe waves and sound wave propagating in the same direction as the probe waves.

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to SBS. The wave vector of the acoustic wave $\vec{q}$ should satisfy the Bragg's condition (momentum conservation; see Fig. 2)

$$\vec{k}_2 - \vec{k}_1 = \vec{q}\,, \tag{1}$$

and, by energy conservation, the frequency of the acoustic wave ω_b should satisfy

$$\nu_1 - \nu_2 = \omega_b\,. \tag{2}$$

Condition (1) implies

$$\begin{aligned} |\vec{q}|^2 &= |\vec{k}_1|^2 + |\vec{k}_2|^2 - 2|\vec{k}_1|\,|\vec{k}_2|\cos\theta \\ &= (|\vec{k}_1| - |\vec{k}_2|)^2 + 4|\vec{k}_1|\,|\vec{k}_2|\sin^2\frac{\theta}{2}\,. \end{aligned} \tag{3}$$

For a dispersionless medium with linear index of refraction n the absolute values of the wave vectors $|\vec{k}_1|$ and $|\vec{k}_2|$ almost coincide, i.e., $|\vec{k}_2| \approx |\vec{k}_1| = \nu n/c$, if $\nu_2 \approx \nu_1 = \nu$, and dispersion relation for the sound wave is

$$|\vec{q}| = \omega_b/V_s\,, \tag{4}$$

where V_s is the speed of sound in the material. Then (3) and (4) imply

$$|\vec{q}| = 2\frac{\nu n}{c}\sin\frac{\theta}{2}\,, \tag{5}$$

$$\omega_b = 2\frac{nV_s}{c}\,\nu\sin\frac{\theta}{2}\,. \tag{6}$$

Conditions (5) and (6) show that the forward SBS ($\theta = 0$) vanishes.

In an ultradispersion medium

$$\begin{aligned} |\vec{k}_1| - |\vec{k}_2| &= \frac{\nu_1 n(\nu_1)}{c} - \frac{\nu_2 n(\nu_2)}{c} \\ &\approx \frac{\nu_1 - \nu_2}{c}\frac{\partial[\nu n(\nu)]}{\partial\nu} = \frac{\nu_1 - \nu_2}{V_g}\,, \end{aligned} \tag{7}$$

where V_g is the group velocity of light. When $V_g \approx c/n$, as in dispersionless medium, we arrive at the previous results. However, when V_g is small enough we find a new expression for the acoustic wave vector.

In particular, the wave vector difference $|\vec{k}_1| - |\vec{k}_2|$ appearing in Eq. (7) is now nonzero. From (2) and (7) we have $|\vec{k}_1| - |\vec{k}_2| = \omega_b/V_g$. Taking $|\vec{k}_1| = \nu n/c$, and from (7) $|\vec{k}_2| = \nu n/c - \omega_b/V_g$, we can write Eq. (3) as

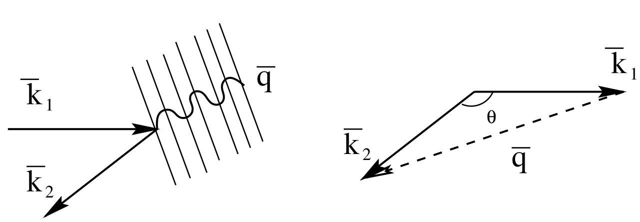

FIG. 2. Geometry for Brillouin scattering.

$$|\vec{q}| = \sqrt{\left(\frac{\omega_b}{V_g}\right)^2 + 4\frac{\nu n}{c}\left(\frac{\nu n}{c} - \frac{\omega_b}{V_g}\right)\sin^2\frac{\theta}{2}}\,, \tag{8}$$

and because of (4) Eq. (8) can be used to obtain the equation for the sound frequency

$$\omega_b^2\frac{V_g^2 - V_s^2}{4V_s^2V_g^2} - \frac{\nu n}{c}\left(\frac{\nu n}{c} - \frac{\omega_b}{V_g}\right)\sin^2\frac{\theta}{2} = 0\,. \tag{9}$$

Equations (8) and (9) coincide with (5) and (6) for $V_g \gg V_s$.

The dispersion relation (9) shows that there is no scattering if $V_s > V_g$, because both roots of Eq. (9) are nonpositive ($|\vec{k}_2| > 0$). The acoustic frequency for backward scattering when $V_g \geq V_s$ is

$$\omega_b^{\text{backward}} = 2\frac{nV_s}{c}\,\nu\,\frac{V_g}{V_g + V_s}\,, \tag{10}$$

while for the forward scattering

$$\begin{aligned} \omega_b^{\text{forward}} &\leq \frac{nV_s}{c}\,\nu \quad \text{if } V_g = V_s\,, \\ \omega_b^{\text{forward}} &= 0 \quad \text{if } V_s \neq V_g\,, \end{aligned} \tag{11}$$

which is drastically different from the case of dispersionless medium. For example, when $V_g \to V_s$ the wave vector $|\vec{k}_2|$ goes to zero and, therefore, backward scattering is forbidden. In contrast, forward scattering is strongly allowed.

The phase-matching conditions are necessary, but not sufficient, to guarantee efficient SBS. To find the coupling between the acoustic and light waves we write the equations of motion and continuity for the medium as

$$\rho_0\left(\frac{\partial\vec{v}}{\partial t}\right) = -\vec{\nabla}[p + (\rho - \rho_0)V_s^2]\,, \tag{12}$$

$$\frac{\partial\rho}{\partial t} + \rho_0\,\mathrm{div}\vec{v} = 0\,, \tag{13}$$

where $\vec{v}$ is the velocity of the medium, p is the pressure, $\rho(\vec{r},t)$ is the density of the medium, and ρ_0 is the unperturbed density. The equations are written in lossless linear approximation, and we assume that $\rho_0 \gg |\rho(\vec{r},t) - \rho_0|$.

Using Eqs. (12) and (13) we find

$$\frac{\partial^2\rho}{\partial t^2} - V_s^2\nabla^2\rho - 2\gamma_{\text{ph}}\frac{\partial\rho}{\partial t} = \nabla^2(\Delta p)\,, \tag{14}$$

where the phonon decay rate term going as γ_{ph} has been added "by hand" following the treatment of [1,12]. The pressure deviation in a medium originating from the interaction with electromagnetic fields Δp is given by [1]

$$\Delta p \approx -N\langle U\rangle\,, \tag{15}$$

where N is the density of the particles of the medium interacting with the electromagnetic field, U is the mean interaction energy of the particles and electromagnetic field. The density modulations result in a running diffraction grating described by

$$\Delta\varepsilon = \left(\frac{\partial\varepsilon}{\partial\rho}\right)_S (\rho - \rho_0), \qquad (16)$$

which leads to SBS.

We focus on forward SBS in a gas of three-level atoms in a Λ configuration first (Fig. 1). The present work also applies to a dielectric doped with such atoms/ions. Assuming that the acousto-optic interaction is weak enough, we solve the problem of the interaction of the three-level atoms with the applied electromagnetic fields and derive the corresponding permittivity of the medium. We calculate the amplitude of the medium density deviations using (14), and from (16) derive the equations for the coupling of electromagnetic and acoustic waves, and an effective interaction Hamiltonian between the waves.

For copropagating electromagnetic waves, as indicated in Fig. 1, the interaction Hamiltonian can be written as

$$\begin{aligned} H = \hbar\delta(|c\rangle\langle c| - |b\rangle\langle b|) \\ - \hbar[|b\rangle\langle a|\Omega_p^* + |c\rangle\langle a|\Omega_d^* + \text{adj}], \end{aligned} \qquad (17)$$

where $|a\rangle\langle a|$, $|c\rangle\langle c|$, $|b\rangle\langle a|$, and $|c\rangle\langle a|$ are the atomic projection operators, 2δ is the two-photon detuning, Ω_p and Ω_d are the probe and drive Rabi frequencies (i.e., $\Omega_p = \wp_{ab}E_p/\hbar$, etc.), $\wp_{ab}$ and $\wp_{ac}$ are the dipole momenta of the transitions. The drive field is resonant with $|a\rangle \to |c\rangle$ transition as per Fig. 1.

Analytic expressions for the expectation values of the lowering operators of the atom σ_{ba} and σ_{ca} can be obtained from the stationary solution of the Bloch equations for the atomic populations and polarizations. The solution is

$$\sigma_{ba} = \Omega_p\left(\frac{i\gamma_0}{|\Omega|^2} - \frac{2\delta|\Omega_d|^2}{|\Omega|^4}\right), \qquad (18)$$

$$\sigma_{ca} = \Omega_d\left(\frac{i\gamma_0}{|\Omega|^2} + \frac{2\delta|\Omega_p|^2}{|\Omega|^4}\right), \qquad (19)$$

$$\sigma_{cc} = \frac{|\Omega_p|^2}{|\Omega|^2}, \qquad \sigma_{bb} = \frac{|\Omega_d|^2}{|\Omega|^2}, \qquad \sigma_{aa} = 0. \qquad (20)$$

where γ_0 is the decay rate of the ground state coherence, $|\Omega|^2 = |\Omega_p|^2 + |\Omega_d|^2 \gg \gamma_0\gamma_r$, γ_r is the homogeneous linewidth of the transitions $|a\rangle \to |b\rangle$ and $|a\rangle \to |c\rangle$. Considering the case of $\gamma_0 \to 0$, and using Eqs. (17)–(20), we obtain an approximation of the expectation value of the Hamiltonian

$$\mathrm{Tr}(H\sigma) = \langle H\rangle \simeq \hbar\delta\left[\frac{|\Omega_p|^2}{|\Omega|^2} - \frac{|\Omega_d|^2}{|\Omega|^2}\right] + O(\delta^3). \qquad (21)$$

To find the acoustic density wave ρ we use the fact that $U \equiv \langle H\rangle$, and from Eq. (14) we see that the force driving the acoustic wave goes as $\partial^2\langle H\rangle/(\partial z^2)$. We envision a constant (cw) drive E_d and probe E_p. The probe consists of two fields of frequencies ν_1 and ν_2 that lay in the EIT transparency gap, that is $|\Omega|^2/\gamma_r \gg \delta \gg |\nu_1 - \nu_2|$, where $|\Omega|^2/\gamma_r$ is the width of the EIT window

$$E_p = E_{p1}e^{-i(\nu_1 t - k_1 z)} + E_{p2}e^{-i(\nu_2 t - k_2 z)}.$$

Assuming a solution of Eq. (14) of the form

$$\rho - \rho_0 = -i\frac{q}{v}\sqrt{\frac{\hbar m}{2\omega_b}}\left(be^{-i(\omega_b t - qz)} - b^\dagger e^{i(\omega_b t - qz)}\right), \qquad (22)$$

where b and $b^\dagger$ are the creation and annihilation operators of the phonon field, v is the total volume of the sample, $m = \rho_0 v$ is the mass of the sample; and using (15) and (21) we arrive at an expression describing acousto-optical interaction in the dielectric

$$\dot{\tilde{b}} + (i\Delta\omega + \gamma_{\mathrm{ph}})\tilde{b} = q\delta N v\sqrt{\frac{2\hbar}{m\omega_b}}\frac{\Omega_{p1}\Omega_{p2}^*}{|\Omega_d|^2}, \qquad (23)$$

where $\tilde{b} = b\exp(-i\Delta\omega t)$, $\Delta\omega = \omega_b - \nu_1 + \nu_2$ [13].

The intensity of the drive field is not a free parameter. To fulfill phase matching conditions (11) the group velocity of light should equal the speed of the sound, that is

$$V_s = V_g = \frac{8\pi|\Omega_d|^2}{3N\lambda^2\gamma_r}, \qquad (24)$$

where $\lambda = 2\pi c/\nu$ is the wavelength of the laser fields, and $n \approx 1$ [14]. Presenting the probe fields as

$$E_{pj} = \sqrt{\frac{2\pi\hbar\nu}{v}}a_{pj}e^{-i(\nu_j t - k_j z)}, \qquad j = 1, 2, \qquad (25)$$

where a_{pj} is the annihilation operator we rewrite Eq. (23) as

$$\dot{\tilde{b}} + (i\Delta\omega + \gamma_{\mathrm{ph}})\tilde{b} = \nu\sqrt{\frac{2\hbar\delta^2}{mV_s^2\omega_b}}\frac{q}{k}a_{p2}^\dagger a_{p1}. \qquad (26)$$

To find the equations describing coupling of the probe fields to the acoustic wave we use the facts that $\varepsilon - 1 = 4\pi N\wp_{ab}\sigma_{ab}/E_p$ and $(\partial N/\partial\rho)_S = N/\rho_0$. Then, using (18), we derive

$$\left(\frac{\partial\varepsilon}{\partial\rho}\right)_S = -\frac{4\pi}{\hbar}\frac{N(2\delta - i\gamma_0)}{\rho_0}\frac{\wp_{ab}^2}{|\Omega_d|^2}. \qquad (27)$$

Taking into account Eqs. (16) and (22) and applying the method of slowly varying amplitudes to equation $\dot{E}_p(t) = i\nu\Delta\varepsilon(t)E_p(t)/2$, we obtain the set of equations for the probe fields

$$\dot{a}_{p1} = -\gamma_0\frac{c}{V_s}a_{p1} - \nu\sqrt{\frac{2\hbar\delta^2}{mV_s^2\omega_b}}\frac{q}{k}a_{p2}\tilde{b}, \qquad (28)$$

$$\dot{a}_{p2} = -\gamma_0\frac{c}{V_s}a_{p2} + \nu\sqrt{\frac{2\hbar\delta^2}{mV_s^2\omega_b}}\frac{q}{k}a_{p1}\tilde{b}^\dagger, \qquad (29)$$

where we assume that $\delta \gg \gamma_0$. The linear dispersion term appearing due to Λ atoms, and proportional to the two-photon detuning, is already taken into account in wave vectors $\vec{k}_1$ and $\vec{k}_2$ and is not included in (28) and (29). Thus the nonlinear interaction between the fields and phonons can be described by the Hamiltonian

$$H_0 = -i\hbar g(a_{p1}^\dagger a_{p2} b - b^\dagger a_{p2}^\dagger a_{p1}), \qquad (30)$$

with coupling constant

$$g = \nu \sqrt{\frac{2\hbar\delta^2}{mV_s^2\omega_b}}\,\frac{q}{k}\,. \qquad (31)$$

It is especially interesting to consider the light propagation through an atomic cell and study the sound waves, generated via the acousto-optic interaction. If a cell with volume $v = 5$ cm^3 contains atomic vapor (mass per atom $m_a \approx 10^{-22}$ g) with density of the three level atoms $N = 10^{12}$ cm^{-3} and density of a buffer gas 10^{18} cm^{-3}, the coupling constant (31) for $\delta = \omega_b = 10^6$ s^{-1} and $V_s = 3\ 10^4$ cm/s is $g \approx 1$ s^{-1}. Minimum group velocity of light in an atomic cell with length $2L$ can be estimated as $V_g \approx 2\gamma_0 L$, and for $L = 1$ cm and $\gamma_0 = 1.5\ 10^4$ s^{-1} we have phase matching $V_s = V_g$ and photon-phonon interaction can be realized in practice.

To estimate the value of the coupling constant for a doped solid we take $\nu = 10^{15}$ s^{-1}, $m = 3\,10^{-4}$ g (which corresponds to a fiber with cross sectional area $\mathcal{A} = 10^{-7}$ cm^2 and length $L = 10^3$ cm), $V_s = 10^5$ cm/s, $\delta = \omega_b = 10^8$ s^{-1}. We get $g \approx 8$ s^{-1}, as in fused silica [8].

As a simple experimental test of the effect we propose to inject monochromatic probe wave a_{p1} into the cell with externally driven sound wave and look for the modulation of the probe. That is, the acoustic wave initiates generation of Stokes a_s and anti-Stokes a_{as} harmonics of the probe field. The corresponding interaction Hamiltonian is similar to (30) and reads

$$H_1 = -i\hbar g(a_{p1}^\dagger a_s + a_{as}^\dagger a_{p1})b + \text{adj}\,. \qquad (32)$$

If the intensity of the phonon field is large enough and the probe is weak enough we can replace the operator b in (32) by the constant c-number B. In such a case the set of equations generated by (32) with appropriate decay terms yields

$$a_{s,as} = \pm\frac{1}{\sqrt{2}}\,a_{p1}(0)e^{-\gamma_0 z/V_s}\sin\sqrt{2}\,g|B|\,\frac{z}{c}\,. \qquad (33)$$

For an acoustic wave with power $P_s = 10$ nW/cm^2 the amplitude B is determined by $|B|^2 = P_s v/(\hbar\omega_s V_s) \simeq 2\,10^{16}$. Inserting this value of B into (33) and taking $g \approx 1$ s^{-1} as per the discussion we find $a_{s,as}(2L) \approx \pm 0.003 a_{p1}(0)$. This should be really observable and provides a direct test of the present effect.

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[13] For monochromatic probe and drive fields there is no acousto-optical interaction, as is seen from (21). To achieve the nonlinear interaction, the effect of EIT should be partially destroyed. This is similar to the conclusion of [15], where it has been shown that large refractive nonlinearity based on the atomic coherence can be achieved only for a "disturbed" EIT regime. An ideal EIT medium itself does not interact with the light [16].
[14] When instead of Λ atoms we use far detuned two-level atoms, the phase-matching conditions are fulfilled for the detuning comparable with Ω_d, if the other conditions are the same (dispersions in both cases should coincide). This essentially means that the absorption due to spontaneous decay of the two-level system is much larger than that of the three-level system.
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