Phase conjugation in stimulated resonant scattering of light with a small frequency shift

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During the excitation of Rb vapor mixed with a quenching buffer gas (N_2), at a deviation of 20–50 cm^{-1} from resonance, a stimulated backscattering with a frequency shift $\sim$0.05–0.4 cm^{-1} has been attained. A phase conjugation of the pump wave has been observed.

An amplification of a weak light wave E by a two-level system in the presence of an intense laser wave E_L, which determines the possibility of a stimulated resonance scattering of light, was predicted theoretically by Rautian and Sobel'man[1] and has subsequently been studied in several places, e.g., Refs. 2 and 3. An exchange of energy between light waves arises because of a modulation of the populations of atomic levels at the difference frequency of the waves. At a deviation from resonance which is large—considerably greater than the width of the atomic transition, $\Delta\omega_0$—one of the two gain maxima lies at the frequency of three-wave mixing and results from a coincidence of the modulation frequency with the Rabi oscillation frequency. The second maximum is characterized by a very small shift with respect to the pump frequency ω_L. This second maximum arises when the modulation frequency is equal to the reciprocal of the time scale of the relaxation induced by the field of the grating of populations. A stimulated resonant scattering at a frequency close to ω_L has been observed[4] during excitation in a ring resonator containing a cell holding Na vapor. Emission arose in the resonator only in a direction making a small angle with the direction of the pump beam.

The excitation of stimulated resonant backscattering with a small frequency shift in metal vapor is hindered by the disruption of the small-scale population grating by the thermal motion of the atoms. The theoretical estimates below show that it is possible to achieve a high gain for stimulated resonant scattering by adding a quenching buffer gas to the vapor. This buffer gas is to shorten the population relaxation time T_1 and to reduce the mean free path of the atoms, $\bar{l}$.

The susceptibility at the frequency of the weak wave, ω, found from the solution of the equations for the elements of the density matrix, with interdiffusion of the excited and unexcited atoms, consists of two parts: $\chi = \chi_a + \chi_b$, where χ_b is unrelated to the population modulation, while χ_b reflects the effect of this modulation. If the Doppler frequency shift is ignored (we will take this shift up below), the imaginary part, χ_a'', is

$$\chi_a'' = -\frac{N\mu^2}{\hbar T_2}\,\frac{1}{(1+S)(\Delta^2 + T_2^{-2})}\,. \tag{1}$$

Here N is the density of atoms, μ is the dipole-moment matrix element, $T_2 = 2/\Delta\omega_0$, $S = (T_1/T_2)\zeta$, $\zeta = \mu^2|E_L|^2/[\hbar^2(\Delta_L^2 + T_2^{-2})]$, $\Delta_L = \omega_L - \omega_0$, ω_0 is the transition frequency, and $\Delta = \omega - \omega_0$. The statistical weights of the levels are assumed to be identical. The time T_1 is given by the expression $T_1^{-1} = w_q + rA_{21}$, where w_q is the probability for the quenching of the excited atom, A_{21} is the probability for a spontaneous decay, and the factor $r < 1$ incorporates the reabsorption of photons of the spontaneous emission.

Under the conditions $\rho = 2|\Delta_L|\Delta\omega_0 \gg 1$ and $|\Omega| \ll |\Delta_L|$, where $\Omega = \omega - \omega_L$, the imaginary part χ_b'' is given by

$$\chi_b'' = -\frac{N\mu^2}{\hbar\,\Delta_L}\,\frac{S}{(1+S)(\alpha+S)}\,\frac{f\xi - \rho^{-1}}{1+\xi^2}\,. \tag{2}$$

Here $\alpha = 1 + (T_1/\tau_d)$, τ_d is the time of the disruption of the population grating by the motion of the atoms, $f = 1 - \Omega_g/\Delta\omega_0$, $\Omega_g = [(\alpha + S)/T_1(1+\zeta)]$ (Ω_g^{-1} is the relaxation time of the population grating after the removal of a weak field), and $\xi = \Omega/\Omega_g$. If τ is much smaller than the grating period Λ, we have $\tau_d = (1/q^2D) \sim p$, where D is the diffusion coefficient, $q = 2\pi/\Lambda$, and p is the pressure of the buffer gas. At $\bar{l} > (\Lambda/2)$ ($p < \hat{p}$, where $\hat{p}$ is the pressure corresponding to $\bar{l} = \Lambda/2$), we have $\tau_d \approx \hat{\tau} = (\Lambda/2\bar{v})$, where $\bar{v}$ is the average velocity of the atoms. Expression (2) ignores the relaxation of the population grating due to the radiative transport of excitation; this simplification is legitimate if at least one of the two conditions $T_1 \ll A_{21}^{-1}$ and

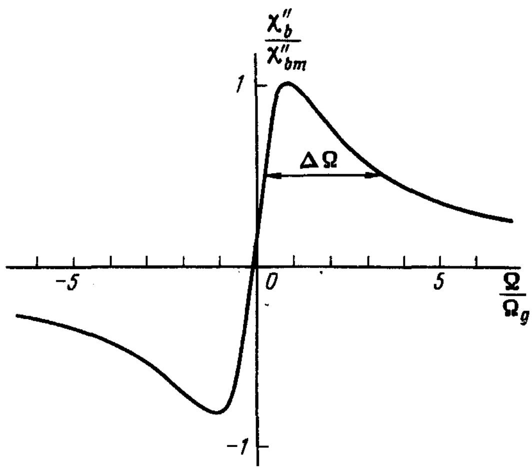

FIG. 1. Dependence of χ''_b on the frequency shift $\Omega = \omega - \omega_L$ ($\rho f = -10$).

$\tau_r \gg |\Omega|^{-1}$ holds, where τ_r is the time scale for the "diffusion" of the spontaneous emission over a distance $\Lambda/2$.

If $|\rho|^{-1} \ll |f|$, then χ''_b reaches its maximum positive value χ''_{bm} at a frequency shift $|\Omega_m| = \Omega_g$, whose sign is determined by the condition $\Omega_m f \Delta_L < 0$ (Fig. 1).

An amplification of the field E is possible if $\chi''_{bm} + \chi''_a > 0$ and $\eta = (\chi''_{bm} / - \chi''_a) > 1$. It follows from (1) and (2) that we have $\eta = \frac{1}{2}|f||\rho| S(\alpha + S)^{-1}$. At $\eta \gg 1$, the width of the gain band is $\Delta\Omega \approx 2\sqrt{3}\,\Omega_m$ (Fig. 1). The distribution of moving atoms in the frequency $\omega_m = \omega_L + \Omega_m$, which is characterized by twice the Doppler width, $2\Delta\Omega_D$, in the case of backscattering, does not reduce the gain if the condition $\Delta\Omega \gg 2\Delta\omega_D$ holds.

The gain for the field E at the frequency ω_m is $\omega_m\ g_m = g_{bm}\ (1 - \eta^{-1})$, where $g_{bm} = (4\pi\omega_m / c)\ \chi''_{bm}$. This gain is related to the absorption coefficient for the pump wave, $g_L \approx (4\pi\omega /_L c)|\chi''_a|$, by $g_m = g_L(\eta - 1)$. The excitation of stimulated resonant scattering in a single pass through the medium requires high values of g_m and also large values of η, in order to reduce the absorption of the pump. Since the relation $g_{bm} \sim (1/\Delta_L)$ holds, and we have $\eta \sim \Delta_L$, the possibility of satisfying these requirements simultaneously through a suitable choice of Δ_L depends on the quantity $Q = g_{bm}\ \eta \approx g_m\ \eta$, which does not depend on Δ_L if $S = \text{const}$.

Depending on the sign of f, one draws a distinction between two regimes of stimulated resonant scattering, in which the frequency shifts are in opposite directions. In the first regime ($f > 0$) the maximum values of Q, which are $(2.5\text{–}5) \times 10^3\ \text{cm}^{-1}$ for resonant transitions of alkali metals, are reached at high values of p ($T_1 \ll \tau_d$, $\alpha \sim 1$), $S \sim 2$, and high values of N, at which the resonant broadening $\Delta\omega'_0$ dominates $\Delta\omega_0$ [($\gamma = (\Delta\omega'_0 / \Delta\omega_0) \sim 1$; with α and $S = \text{const}$, we have $Q \sim \gamma$]. Since we have $\Delta\omega_D \approx 2.3/\hat{\tau}$ and $\Delta\Omega > (2\sqrt{3}/T_1)$, the Doppler frequency shift has only a slight effect at $T_1 \ll \hat{\tau}$.

The second regime ($f<0$) prevails at $p \ll \hat{p}$ and modest values of N, at which T_2 is much longer than $\hat{\tau}$. The maximum values of Q, which are reached at $\alpha \gg (T_1/T_2)$, $S \sim (T_1/T_2)$, $\gamma \sim 1/2$, are lower than those given above by a factor $\sim 5(T_1/T_2)$. The reason for the high values of Q in the first regime is that under the condition $T_1 \ll \tau_d$ the motion of the atoms does not manage to disrupt the population grating. In the second regime, in contrast, the motion of the atoms is the primary factor in the relaxation of this grating.

If the saturation of the medium is slight ($S \ll 1$), we have $g_m(\mathbf{r}) \sim |E_L(\mathbf{r})|^2$; this relation tells us that the phase conjugation of a spatially nonuniform pump is possible.

In the experiments we used a dye laser with a pulse length of 15 ns and an output line of width 10^{-2} cm^{-1}, whose frequency could be tuned near the resonant line Rb $5^2S_{1/2} - 5^2P_{3/2}$ ($\lambda = 780$ nm). A laser beam 4 mm in diameter was focused into a cell by a lens with $f = 9$ cm at a distance $l_f = 3$ cm from the entrance window. A phase plate could be placed in the path of the pump beam to increase its angular divergence from 1 to 8 mrad. The N_2 pressure in the cold cell was 0.8 atm.

At temperatures of 310–340 °C ($N = 2.9 \times 10^{16}$–5.2×10^{16} cm^{-3}) and at deviations $\Delta_L \approx -40$ cm^{-1} from resonance, the stimulated resonant backscattering arose at a pump power $P_L \sim 100$–150 kW. At $P_L \sim 300$–400 kW, the power of the backscattering was $\sim 0.01 P_L$. The stimulated-resonant-backscattering line was on the short-wave side of ω_L (Fig. 2). The length of the stimulated-resonant-backscattering pulse was 7–8 ns, and its peak coincided with the peak of the pump pulse. In operation with a phase plate we observed a phase conjugation of the pump (although not stably). After passing through the phase plate, the stimulated resonant backscattering reconstructed the original angular divergence of the pump.

From the absorption of the vapor at various intensities, deviations from resonance, and temperatures, we found $T_1 \sim 0.15$ ns and $T \sim 0.02$ ns. Using the estimate $\tau_d \sim 0.5$ ns, we find $\alpha \sim 1.3$ and $f \approx 1$ (for the first regime of stimulated resonant scattering). Estimates from (1) and (2) at the threshold values of P_L yield $g_L \sim 1$ and $g_{bm} \gtrsim 10$, which would be sufficient, in view of the value of l_f, for the excitation of the stimulated resonant scattering from spontaneous-scattering "noise."

When we tuned the pump frequency, we observed a stimulated resonant scattering in the interval from $\Delta_L \approx -20$ cm^{-1} to $\Delta_L \approx -50$ cm^{-1}. It was excited in approximately the same way in the region of short-wave deviations ($\Delta_L > 0$); the

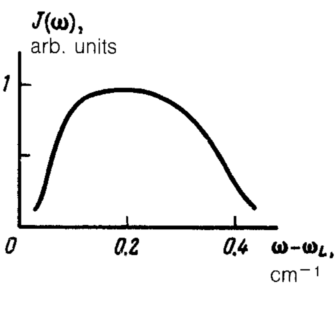

FIG. 2. Experimental shape of the spectral line of stimulated resonant backscattering, $J(\omega)$ ($P_L = 400$ kW, $\Delta_L = -40$ cm^{-1}, $t = 330$ °C).

direction of the frequency shift was reversed. The stimulated resonant scattering was also excited, although at a lower intensity, when the spectral width of the laser was 0.3 cm^{-1}. It did not arise when this width was increased to 2 cm^{-1}.

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