Coherent Raman response and spectral characteristics of ultrashort solitons in fibers

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The Raman self-frequency shift of a soliton is studied for pulse durations shorter than the optical phonon oscillation period and with respect to the influence of the field-induced change in the vibrational level populations. It is shown that under certain conditions the self-frequency shift can be suppressed by the coherent saturation of the Raman response. [S1063-651X(96)09805-4]

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I. INTRODUCTION

Since the theoretical prediction [1] and experimental observation [2] of solitons in single-mode fibers, this phenomenon has been the continual focus of both experimental and theoretical investigations. In recent years much attention has been paid to the analysis of the influence of a variety of physical processes on the soliton dynamics. Among these the effect of stimulated Raman scattering (SRS) can play an essential role in the propagation of solitons [3–9]. This process generally leads to the breakup of multisoliton bound states [5–7]. In contrast, the structure of a fundamental soliton turns out to be quite stable. However, the Raman effect gives rise to a continuous downshift of the soliton carrier frequency [the Raman self-frequency shift (RSFS) [3]]. The theory of the RSFS predicts a frequency shift depending on the inverse fourth power of the soliton duration. The appearance of such effects is caused by the broad Raman line in fibers extending down to zero detuning.

However, in recent experiments on soliton transmission and amplification in fibers, pulses with durations as short as 20 fs were observed [10–12]. Since the optical phonon oscillation period in fibers (corresponding to the Stokes shift of 440 cm^{-1}) is about 75 fs, in these experiments for the pulse duration t_p, the condition

$$t_p < 2\pi\Omega_R^{-1} \tag{1}$$

is fulfilled where $\hbar\Omega_R = E_2 - E_1$ is the energy difference between the Raman vibrational levels.

The RSFS under such conditions shows some qualitatively new features [13]. First, the structure of the Raman gain line no longer determines the characteristics of SRS of the soliton. Now the process of intrapulse SRS occurs because the initial spectrum of the pulse contains already both pump and Stokes components comparable in amplitude and satisfying the condition of SRS resonance. Although in this case the soliton spectral width exceeds the Raman gain width γ_R, the RSFS was shown to further increase with a decrease of the soliton duration. Since the soliton intensity scales as the inverse second power of the soliton duration, one can expect that a field-induced variation of the Raman level population may play an important role in the medium response leading to saturation effects similar to those arising under coherent interaction of quantum systems with intense resonant field [16].

In the present paper we analyze the RSFS for soliton durations smaller than the characteristic period of the lattice vibrations [when the condition (1) is satisfied], taking into account the population change of the vibrational levels. In the limit of long soliton durations the generalized theoretical model yields the results consistent with the results of previous works [8,9]. For soliton durations short compared to the phonon vibrational period in an intermediate region where the population change of the Raman transition is still negligible the RSFS is shown to be inversely proportional to the soliton duration. The RSFS in this parameter region we already studied in Ref. [13]. However, as remarked above, for even shorter and more intensive solitons the coherent dynamics of the population of the Raman levels has to be taken into account. As a result of the investigation in the present paper we find for small pulse durations due to the nutations of the population of the vibrational levels a decreasing self-frequency shift with decreasing pulse duration and under certain conditions a self-induced suppression of the RSFS.

The paper is organized as follows. In Sec. II the basic equations for the laser field amplitude, the normal coordinate, and the population difference of the Raman oscillator are considered. The solution for the Raman oscillator and the Raman response function are analyzed in the different parameter regions for the pulse duration. In Sec. III the soliton self-frequency shift is calculated with the help of the soliton perturbation theory [18]. The results are analyzed and explained by the combined Stokes and anti-Stokes shift under the specified considered conditions.

II. BASIC EQUATIONS

In order to investigate the effect of SRS on the soliton evolution, we start from the semiclassical approach in which the evolution of the pulse field

$$\mathcal{E}(z,t) = \tfrac{1}{2}E(z,t)\{\exp[i(\omega t - kz)] + \text{c.c.}\}$$

is described by the model of a Raman-active two-level quantum oscillator [14] with a characteristic frequency Ω_R, a normal coordinate Q, a population difference between the two levels $\rho = \rho_1 - \rho_2$, and with phenomenological popula-

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tion and polarization relaxation times T_1 and T_2. Note that in the commonly used approximation neglecting a field-induced variation of the populations ($\rho\approx\rho_0=\text{const}$) the model turns into a conventional Lorentz oscillator with an effective mass M excited by a driving force proportional to $\mathscr{E}^2$ with the proportionality factor $(\partial\alpha/\partial Q)$, where $\alpha(Q)$ is the electron polarizability of the Raman transition.

Taking into account the contribution to the medium polarization due to the electronic Kerr effect with the nonlinear index change $\Delta n=\frac{1}{2}n_2|E|^2$, the group velocity dispersion $k_{\omega\omega}=\partial^2k/\partial\omega^2$, and the Raman contribution $P_{\text{Ram}}=N(\partial\alpha/\partial Q)Q\mathscr{E}$ (N is the number density of Raman active oscillators), the following set of normalized equations for the slowly varying field amplitude A, the normal coordinate $\widetilde{Q}$, and the population difference of the Raman oscillator $\widetilde{\rho}$ can be derived:

$$i\frac{\partial A}{\partial\zeta}=\frac{\partial^2A}{\partial\tau^2}+2|A|^2A+\mu\widetilde{Q}A, \tag{2}$$

$$\frac{\partial^2\widetilde{Q}}{\partial\tau^2}+\frac{2}{\tau_2}\frac{\partial\widetilde{Q}}{\partial\tau}+\widetilde{Q}=\rho|A|^2, \tag{3a}$$

$$\frac{\partial\widetilde{\rho}}{\partial\tau}+\frac{\widetilde{\rho}-1}{\tau_1}=-\kappa^2\frac{\partial\widetilde{Q}}{\partial\tau}|A|^2. \tag{3b}$$

In Eqs. (2) and (3), the normalized time $\tau=(t-z/v_g)\Omega_R$, the normalized longitudinal and transversal relaxation times $\tau_{1,2}=T_{1,2}\Omega_R$, the normalized propagation distance $\zeta=z/z_d$ with $z_d=2|k_{\omega\omega}|^{-1}\Omega_R^{-2}$, the normalized field amplitude $A=(E/E_0)$, $E_0=(2n_0|k_{\omega\omega}|\Omega_R^2/kn_2)^{1/2}$, the vibrational normal coordinate $\widetilde{Q}=(Q/Q_0)$, $Q_0=|k_{\omega\omega}|n_0\rho_0/2Mkn_2(\partial\alpha/\partial Q)$, and the population difference $\widetilde{\rho}=(\rho/\rho_0)$ were introduced.

Since we consider here the process of intrapulse SRS, pump and Stokes frequency components of the pulse spectrum are described in Eqs. (2) and (3) by a single time-dependent pulse amplitude.

The field-induced change in the population of the Raman levels is characterized by the parameter

$$\kappa^2=\left(\frac{MQ_0^2\Omega_R^2}{\rho_0^2\hbar\Omega_R}\right)=\tfrac{1}{4}(\hbar\Omega_RM)^2\left(\frac{\partial\alpha}{\partial Q}\right)^2, \tag{4}$$

while the coupling between the oscillator and the pulse field is described by

$$\mu=\frac{2\pi N\rho_0\Omega_R^{-2}}{n_0n_2M}\left(\frac{\partial\alpha}{\partial Q}\right)^2. \tag{5}$$

Equations (3) should be supplemented by the initial conditions

$$\widetilde{Q}(z,-\infty)=\frac{\partial\widetilde{Q}}{\partial\tau_{(z,-\infty)}}=0,\quad \widetilde{\rho}(z,-\infty)=1. \tag{6}$$

Note that the terms describing nonlinear and dispersive effects of higher order with respect to the small parameter $(2\pi/\omega t_p)$ are omitted in Eq. (2), since these make no contribution to the continuous frequency shift [15].

At first we check the model (3) in the limit of long pulses $\tau_p=\Omega_Rt_p\gg1$. Indeed, the limit of long durations corresponds to relatively small soliton intensities, and one can neglect the variation of $\widetilde{\rho}$ in Eq. (3). Then the general solution of Eq. (3a) can be written using the casual response function

$$\widetilde{Q}(\tau)=\int_0^\infty d\ sf(s)|A(\tau-s)|^2, \tag{7a}$$

where

$$f(s)=-(1/\omega_R)\exp\{-\gamma_Rs\}\sin(\omega_Rs), \tag{7b}$$

$$\omega_R=(1-\gamma_R^2)^{1/2},\quad \gamma_R=\tau_2^{-1}.$$

The Raman response (7a) can be formally represented by the series

$$\widetilde{Q}(\tau)=-\text{Im}\left(\frac{1}{\omega_R}\sum_{k=0}^{\infty}\frac{(-1)^{k+1}}{[\gamma_R+i\omega_R]^{k+1}}\frac{\partial^k}{\partial\tau^k}|A(\tau)|^2\right). \tag{8}$$

For a pulse field $A(\tau)$ with a characteristic time scale τ_p, Eq. (8) can be considered as an expansion with the parameter $\delta\sim\tau_p^{-1}|\gamma_R+i\omega_R|^{-1}$, which becomes small for pulse durations $\tau_p\gg1$.

Taking into account only the first two terms of the series (8) for pulses with the durations $t_p>T_R$, we have

$$\widetilde{Q}(\tau)=|A(\tau)|^2-2\gamma_R\frac{\partial}{\partial\tau}|A(\tau)|^2, \tag{9}$$

where the first term contributes to the medium Kerr nonlinearity, while the second one leads to the RSFS of a soliton proportional to τ_p^{-4}, in agreement with the results of Refs. [8,9].

The situation changes significantly in the case of pulses with small durations characterized by the relation (1) or $\tau_p\ll1$. In this case the second and third terms in Eq. (3a) and the second term in Eq. (3b) can be neglected, and we have to solve the following equations:

$$\frac{\partial^2\widetilde{Q}}{\partial\tau^2}=\widetilde{\rho}|A|^2, \tag{10a}$$

$$\frac{\partial\widetilde{\rho}}{\partial\tau}=-\kappa^2|A|^2\frac{\partial\widetilde{Q}}{\partial\tau}. \tag{10b}$$

Taking into account the initial conditions (6), we can find the solution

$$\frac{\partial\widetilde{Q}}{\partial\tau}=\kappa^{-1}\sin\Psi(\tau),\quad \widetilde{\rho}=\cos\Psi(\tau), \tag{11}$$

where we introduced the variable $\Psi(\tau)=\kappa\int_{-\infty}^{\tau}|A(\tau')|^2d\tau'$ proportional to the instantaneous pulse energy.

The normal coordinate of the oscillator then is given by the expression:

$$\widetilde{Q}(\tau)=\kappa^{-1}\int_{-\infty}^{\tau}d\tau'\sin\left\{\kappa\int_{-\infty}^{\tau'}d\tau''|A(\tau'')|^2\right\}. \tag{12}$$

Unlike the case of long-pulse SRS described by Eq. (9), the normal coordinate (12) is a nonlinear function of the instantaneous pulse energy. For pulses with $\Psi(\infty) = \kappa\int|A(\tau)|^2 d\tau > 2\pi$ both the vibrational coordinate and the population difference become oscillating functions of the pulse energy. The above solution for the Raman response describes in fact the coherent nutations of the Raman transition similar to the Rabi nutations in a two-level system subjected to a resonant field [16]. The quantity κ^{-1} then characterizes the normalized coherent saturation energy of the system, $W_{\mathrm{sat}} = \kappa^{-1}$, and from Eq. (12) it follows that for pulses with an energy $W_p = \int|A(\tau)|^2 d\tau > W_{\mathrm{sat}}$ and a characteristic duration τ_p the magnitude of $\tilde{Q}(\tau)$ is limited by the value $-\tau_p\kappa^{-1}$.

III. SOLITON SELF-FREQUENCY SHIFT

Let us now examine the RSFS of a soliton with the Raman response (12). Since far from the saturation the dependence of the Raman response (12) on the pulse parameters can be estimated by $\tilde{Q} \sim \tau_p^2|A_0|^2$ (A_0 is the peak amplitude of the pulse field), the maximal relative contribution of SRS to the medium nonlinearity [see Eq. (2)] is determined by the parameter μ, which can be expressed in terms of the Raman gain $g_R(0)$ in the line center: $\mu = 2n_2 g_R(0)/kn_0\Omega_R T_2$. Using the results of [7,17], for silica glass this parameter can be estimated by $\mu \approx 0.2$, and thus the Raman polarization still may be considered sufficiently small compared with the Kerr electronic effect. This means that the effect of SRS on the soliton evolution can be investigated in the framework of the adiabatic perturbation theory.

Therefore we can represent the solution of Eq. (2) in the form of a fundamental soliton $A_0(\zeta,\tau) = 2\beta \exp(i\theta)\mathrm{sech}(x)$ with the slowly varying parameters $x = 2\beta(\zeta)[\tau - \xi(\zeta)]$, $\theta = 2\gamma(\zeta)[\tau - \xi(\zeta)] + \delta(\zeta)$, and the normalized duration $\tau_p^* = (\tau_p/1.76) = (2\beta)^{-1}$. Then the frequency shift is defined as the time derivative $\Delta\tilde{\omega} = \partial\theta/\partial\tau = 2\gamma(\zeta)$ with [18]:

$$\frac{d\gamma}{d\zeta} = -\tfrac{1}{2}\ \mathrm{Im}\left\{\int dx(i\mu A_0\tilde{Q})\exp(-i\theta)\mathrm{sech}(x)\tanh(x)\right\}. \tag{13}$$

Substituting the expression for the Raman response (12) into Eq. (13) we obtain the following formula for the RSFS:

$$\frac{d\Delta\tilde{\omega}}{d\zeta} = -\frac{1}{2}\mu\frac{\tau_p^*}{\kappa^2}\left[1-\cos\left(\frac{2\kappa}{\tau_p^*}\right)\right]. \tag{14}$$

From (14) one can see that the RSFS of an ultrashort soliton is generally an oscillating function of the inverse soliton duration $(\tau_p^*)^{-1}$ or of the soliton energy $W_p = 2(\tau_p^*)^{-1}$. The time delay $\Delta\tau = \xi(\zeta)$ of the soliton in respect to the unperturbed solution is given by

$$\Delta\tau(\zeta) = \tfrac{1}{2}\mu\frac{\tau_p^*}{\kappa^2}\left[1-\cos\left(\frac{2\kappa}{\tau_p^*}\right)\right]\zeta^2 \tag{15}$$

and, as well as the RSFS, exhibits oscillating dependence on the soliton duration.

If for the soliton duration the condition $\tau_p^* > 2\kappa$ is fulfilled, from relation (14) we find

$$\frac{d\Delta\tilde{\omega}}{d\zeta} = -\mu\left(\frac{1}{\tau_p^*}\right). \tag{16}$$

The process of SRS in this case is still in the linear Lorentz oscillator regime, where the population change can be neglected ($\tilde{\rho} \approx 1$). The RSFS in this intermediate regime ($1 > \tau_p^* > \kappa$) is inversely proportional to the soliton duration [13].

However, in the region $\tau_p^* < \kappa$ the coherent nutations of the vibrational level population come to play a role. From the relation (14) one can see that for discrete values of the soliton duration $\tau_p^*(q)$ [or the energy $W_p(q)$ with $2\kappa/\tau_p^* = 2\pi q$ ($q = 1,2,\ldots$)] the self-frequency shift vanishes. This self-suppression of the RSFS can be understood in the following way. According to Eqs. (10) and (11), within the regions where the value of $\Psi(\tau)$ satisfies the condition $\pi 2q < \Psi(\tau) < \pi(2q+1)$, ($q = 0,1,\ldots$) transitions to the upper level dominate ($\partial\rho/\partial\tau < 0$). These regions correspond to intrapulse Stokes scattering, and the pulse spectrum shifts here towards lower frequencies. Within the regions where $\pi(2q+1) < \Psi(\tau) < \pi(2q+2)$ the population turns back from the upper to the lower level ($\partial\rho/\partial\tau > 0$), and intrapulse anti-Stokes scattering results in the spectral shift towards higher frequencies.

The self-suppression of the RSFS thus can be explained by the fact that at the above specified conditions the red shift of the pulse frequency in the regions of Stokes scattering is completely compensated by the blue shift of its frequency in the regions of anti-Stokes scattering. At these pulse parameters the Raman oscillator is in fact returned by the pulse field in its initial quantum state whereby the ''center of gravity'' of the soliton spectrum remains unchanged.

Let us now estimate the characteristic values of the soliton parameters required for observation of the discussed effects. With the above given estimate for the parameter μ (≈ 0.2) and typical fiber parameters $n_2 = 10^{-16}$ cm^2/W, $k_{\omega\omega} = -3\times 10^{-28}$ s^2/cm, $\Omega_R = 400$ cm^{-1}, $M \approx 10^{-22}$ g, $N \approx 5\times 10^{21}$ cm^{-3}, we find from Eq. (5) that $(\partial\alpha/\partial Q) \approx 2\times 10^{-15}$ cm^2. This gives the Raman saturation energy density in physical units $W_{\mathrm{sat}} \approx 0.1$ J/cm^2.

In Fig. 1 the RSFS is plotted in dependence on the pulse duration with formula (14). For pulse durations smaller than 50 fs with the above given parameters, the RSFS decreases and shows an oscillating behavior with a first zero at $t_p \approx 20$ fs. In curve 2 the RSFS is depicted with formula (14) derived in the linear approximation limit of the Raman oscillator ($\kappa \to 0$). For a comparison in curve 3 the RSFS is depicted using Gordon's formula (8) with a pulse duration dependence of t_p^{-4}. The limit of validity of this formula is restricted for large pulse durations $t_p > T_R \approx 75$ fs. As one can see in the considered time region this formula predicts a value that is remarkably different from our formula (14).

The relation (14) for the RSFS was derived under the specific assumption for solitons in fibers with negative group velocity dispersion (GVD) and using the approximation of

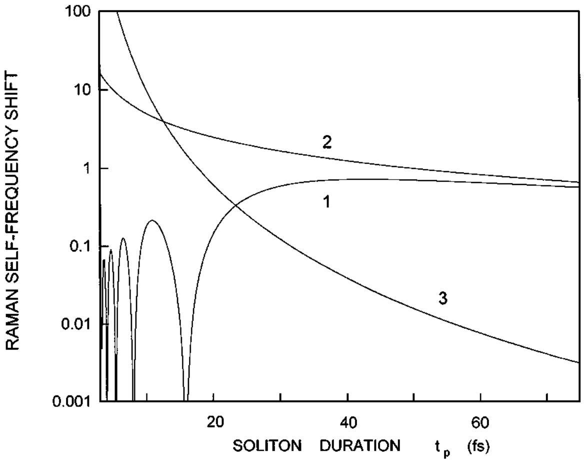

FIG. 1. Normalized soliton of self-frequency shift $(d\Delta\tilde{\omega}/d\zeta)\mu^{-1}$ per unit propagation length as a function of the soliton duration t_p. Curve 1, the RSFS calculated with formula (14) ($W_{sat}=0.1$ J/cm^2); curve 2, the RSFS calculated with formula (14) in the linear approximation limit of the Raman oscillator ($W_{sat}=\infty$); curve 3, the RSFS calculated with the Raman response (9) (Gordon's theory [8]).

soliton perturbation theory. However, a similar but more general relation can be derived for arbitrary short pulses with the condition (1) and without the use of perturbation theory. With a separation of the phase, $A(\zeta,t)=|A|\exp(i\varphi)$, and introducing the mean frequency shift by the notation

$$\langle\dot{\varphi}\rangle=\int_{-\infty}^{\infty}\frac{\dot{\varphi}(\zeta,\tau)|A(\zeta,\tau)|^2d\tau}{\int_{-\infty}^{\infty}|A(\zeta,\tau)|^2d\tau}, \qquad (17)$$

we find from (2) and (11)

$$\frac{d}{d\zeta}\langle\dot{\varphi}\rangle=-\mu W_p^{-1}\int_{-\infty}^{\infty}|A|^2\frac{\partial\tilde{Q}}{\partial\tau}d\tau$$
$$=-\left(\frac{\mu}{\kappa^2}\right)W_p^{-1}[1-\cos(\kappa W_p)], \qquad (18)$$

which holds as well for negative as for positive GVD and an arbitrary peak power. Since for a soliton the pulse energy is given by $W_p=2(\tau_p^*)^{-1}$ the above result reproduces in this specific case the relation (14).

IV. CONCLUSION

In conclusion, we have investigated the effect of stimulated Raman scattering on soliton propagation for solitons with duration smaller than the period of the vibrational oscillations in the medium, taking into account the population change of the vibrational levels. It was shown that the RSFS in this situation gets some qualitatively new features. The frequency shift now is caused not by the broad Raman gain line but rather by the fact that the initial soliton spectrum already contains both pump and Stokes components. In the intermediate region $T_R>t_p>\kappa\Omega_R^{-1}$ the RSFS is inversely proportional to the soliton width and does not depend on the characteristic frequency of the Raman transition and on the Raman gain width. For small pulse durations $t_p<\kappa\Omega_R^{-1}$ due to the nutations of the populations of the vibrational levels the RSFS decreases and shows an oscillating behavior. For the soliton duration $t_p=(2/\pi)\kappa\Omega_R^{-1}$ ($t_p\approx 20$ fs for silica glass) we predict the effect of self-suppression of the continuous downshift of the soliton carrier frequency.

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