Ionization-front soliton in x-ray-stimulated Raman scattering

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We predict solitonlike pulses and precursors at the Stokes frequency in stimulated electronic Raman scattering at the photoionization front of pumping x-ray radiation. We find two different solitonlike patterns that depend on different photoionization mechanisms.

Fast processes in stimulated Raman scattering may result in (bright) soliton formation at the Stokes frequency (combined with the formation of dark[1,2] or bright[3] solitons at the pumping frequency). These formations are true[1] (and substantially coherent whether transient[2] or stable[3]) solitons in the sense that they conserve energy and their duration must be shorter than relaxation times of the underlying two-level Raman-active transition in the material. The emergence of x-ray lasers calls for exploration of new, x-ray nonlinear optical effects.[4] In our most recent research on x-ray stimulated Raman scattering[5] we considered the main new factor: the presence of strong direct or indirect photoionization of the media by both the pump and the Stokes pulses (even at moderate pulse intensities, the x-ray radiation can fully ionize the atoms in a fraction of the pump pulse duration). We showed that, despite an extremely fast photoionization, a significant exponential gain as well as conversion efficiency can be attained in resonantly enhanced stimulated electronic Raman scattering (SERS) in gaseous Li and He. One of the major results is that the presence of SERS can significantly inhibit the photoionization of the media. In this Letter we show also that the x-ray SERS may form stable, solitonlike pulses at the Stokes frequency at the photoionization front of x-ray radiation. To distinguish this phenomenon from the true solitons (whose energy and momentum are conserved in time), we will call it ionization-front stimulated Raman (IFSR) solitons.

In Ref. 5 we proposed to use double-excited intermediate levels of Li and He atoms resonant to a Se^{24+} x-ray laser with a wavelength $\lambda_L = 20.9$ nm. In the case of He, we considered the Raman transition $1\,^1S \rightarrow 2\,^1S$, a Raman wavelength $\lambda_{He} = 59.6$ nm, a double-excited intermediate level $2s2p\,^1P$, and a Stokes wavelength $\lambda_s = 32.2$ nm; in the case of Li, we considered the Raman transition $1s^22s \rightarrow 1s^23s\,^2S$ ($\lambda_{Li} = 239$ nm) and a double-excited intermediate level $[1s(2s2p)^1P]^2P$ ($\lambda_s = 22.3$ nm). We use here a plane-wave approximation and express the pump (E_p) and the Stokes (E_s) fields as $(1/2)E_{p,s}\exp[i(\omega_{p,s}t - k_{p,s}z)] + \text{c.c.}$, where $k_{p,s} = n_{p,s}\omega_{p,s}/c$ and $E_{p,s}$ are slowly varying complex field envelopes. The nondiagonal elements of the Raman transition density matrix, ρ_{jk}, are expressed as $\rho_{21} = \rho_{12}^* = i\rho\exp[i(k_p - k_s)z - i(\omega_p - \omega_s)t]$, where ρ is also a slowly varying envelope.

The photoionization is described by field-dependent decay terms with photoionization coefficients β_j^p and β_j^s ($j = 1, 2$) ascribed to each level j and related to the respective cross sections $\beta_j^{p,s} = \sigma_j^{p,s}\epsilon_0 c/2\hbar\omega_{p,s}$ (the data for $\sigma_j^{p,s}$ can be found in Refs. 6 and 7 for He and Li, respectively). In terms of relaxation rates $w_j = \beta_j^p|E_p|^2 + \beta_j^s|E_s|^2$ and retarded coordinates, $\tau = t - z/c$ and $\xi = z$, the photoionization modified SERS Maxwell–Bloch equations are[5]

$$\partial\rho_{jj}/\partial\tau = (-1)^j K\,\mathrm{Re}(\rho E_p E_s^*) - w_j\rho_{jj} \qquad (j = 1, 2), \tag{1a}$$

$$\partial\rho/\partial\tau = [(\rho_{11} - \rho_{22})KE_p^*E_s - (w_1 + w_2)\rho]/2, \tag{1b}$$

$$\partial E_{p,s}/\partial\xi = (N\hbar\omega_{p,s}/\epsilon_0 c) \times [S_{p,s}\rho K E_{s,p} - E_{p,s}(\beta_1^{p,s}\rho_{11} + \beta_2^{p,s}\rho_{22})]. \tag{1c}$$

Here N is the atomic number density, $S_p = -S_s = -1$, and K is the compound matrix element for the SERS process.[5,8,9] When $K = 0$, Eqs. (1) correspond to the direct photoionization of atoms (from both the levels) by the pump and Stokes radiation with short ionization times, such that the recombination process can be neglected. Equations (1) can be expressed in terms of only photon fluxes $\Phi_{p,s} = |E_{p,s}|^2\epsilon_0 c/2\hbar\omega_{p,s}$ and the envelope ρ, which now becomes purely real.[10] For the stationary propagating solution, all the variables are functions of $\eta = t - z/\nu$ alone, where ν is an (unknown yet) group velocity:

$$\mathrm{d}\rho_{jj}/\mathrm{d}n = (-1)^j\rho U - w_j\rho_{jj} \qquad (j = 1, 2), \tag{2a}$$

$$\mathrm{d}\rho/\mathrm{d}\eta = (\rho_{11} - \rho_{12})U/2 - (w_1 + w_2)\rho/2, \tag{2b}$$

$$\delta\,\mathrm{d}\Phi_i/\mathrm{d}\eta = N[S_i\rho U - (\sigma_1^i\rho_{11} + \sigma_2^i\rho_{22})\Phi_i] \quad (i = p, s), \tag{2c}$$

where $U^2 = \sigma_R{}^2\Phi_p\Phi_s$, $\sigma_R{}^2 = (2K\hbar/\epsilon_0 c)^2\omega_p\omega_s$, $\delta = 1/c - 1/\nu$, and $w_j = \sigma_j^p\Phi_p + \sigma_j^s\Phi_s$. In general, all four photoionization cross sections $\sigma_j^{p,s}$ could be significant. In resonantly enhanced SERS[5] discussed here, there are, however, only two situations: (i) In He the direct photoionization from the ground level

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dominates, i.e., one may assume that $\sigma_2^p \approx \sigma_2^s \approx 0$ [$\sigma_1^p \approx \sigma_1^s \approx 10^{-18}$ cm^2 and $\sigma_1^{p,s}/\sigma_2^{p,s} \gtrsim 100$ (Ref. 6)]. (ii) In Li the indirect photoionization, i.e., absorption to the $[1s(2s2p)^1P]^2P$ level followed by fast autoionization, is the dominant process, i.e., $\sigma_2^p \approx \sigma_1^s \approx 0$.

Let us first consider direct photoionization in the absence of SERS, i.e., when $\Phi_s(\eta) = \rho_{22}(\eta) = \rho(\eta) = U = 0$. Equations (2) are then reduced to

$$d\rho_{11}/d\eta = -\sigma_1^p \Phi_p \rho_{11}, \qquad \delta d\Phi_p/d\eta = -N\sigma_1^p \Phi_p \rho_{11}. \tag{3}$$

The first integral of Eqs. (3) is $\rho_{11} - \delta\Phi_p/N = C$ = constant. Assuming a semi-infinite pump pulse with $\Phi_p(\infty) = \Phi_0$ and boundary conditions $\rho_{11}(\infty) = \Phi_p(-\infty) = 0$ and $\rho_{11}(-\infty) = 1$, we find that $C = 1$ and $\delta = -N/\Phi_0$. Equations (3) can now be integrated exactly: $\rho_{11}(\eta) = (1/2)[1 - \tanh(\eta/\eta_0)]$ and $\Phi_p = \Phi_0(1 - \rho_{11})$, where $\eta_0 = 2(\Phi_0\sigma_1^p)^{-1}$ is the photoionization time. The group velocity ν can thus be identified as the velocity of the photoionization front:

$$\nu = \nu_{\text{ion}} = (1/c + N/\Phi_0)^{-1}. \tag{4}$$

For a pump intensity of 10^{13} W/cm^2 (for the existing Se x-ray laser this requires a confocal parameter of ~0.3 cm), $\eta_0 = 2.85$ ps for Li and $\eta_0 = 2$ ps for He. For $N = 10^{19}$ cm^{-3} (corresponding to ~0.4 atm at $T = 300$ K), we have $\nu_{\text{ion}}/c = 0.77$.

Considering now both pump and Stokes fields in Eqs. (2), we start with an IFSR soliton in He, namely, $\sigma_2^{p,s} = 0$. Figure 1 depicts typical numerical solutions for the intensities and populations obtained with spectroscopic data for He. We assumed semi-infinite pump and Stokes pulses with, initially, $\Phi_s \ll \Phi_p$. The stationary solution in this case consists of a stable Stokes pulse traveling with exactly the same velocity as the photoionization front (and the leading edge of the pump). Figure 1 shows that the coherent SERS process significantly inhibits the photoionization of neutral atoms by x-ray radiation, the degree of this inhibition being measured by the population of the excited Raman level in the wake of the Stokes pulse $\rho_{22}(\infty) = \alpha$. This inhibition occurs because a significant portion of neutral atoms ends up being parked at the upper excited level, whose photoionization cross section is very small. Repeated numerical simulations revealed that neither the shape of the IFSR soliton nor α depends on the shape of the initial leading edge of the pump or Stokes pulses. It was also found that α is purely spectroscopic parameter, independent of neither $\Phi_p(\infty) = \Phi_0$ nor N (for the particular SERS in He, $\alpha_{\text{He}} \approx 0.3$). We can find a useful analytical relation between an unknown group velocity of the IFSR soliton, ν, and the parameter α by considering the constant of motion of these equations (for $\sigma_2^{p,s} = 0$), which exists despite the strong photoionization of the medium. Indeed, defining $\rho_\Sigma = \rho_{11} + \rho_{22}$ and $\Phi_\Sigma = \Phi_s + \Phi_p$, one readily verifies that

$$\rho_\Sigma - \delta\Phi_\Sigma/N = \text{const}. \tag{5}$$

With the boundary conditions $\rho_\Sigma(\infty) = \alpha_{\text{He}}$, $\Phi_\Sigma(\infty) = \Phi_0$, and $\rho_\Sigma(-\infty) = 1$, $\Phi_\Sigma(-\infty) = 0$, the IFSR group velocity in He (which is also the modified photoionization-front velocity) is

$$\nu_1 = [1/c + (1 - \alpha_{\text{He}})N/\Phi_0]^{-1}. \tag{6}$$

Thus the IFSR soliton not only inhibits the photoionization but also permits a much faster propagation of the photoionization front as compared with non-Stokes propagation, given by Eq. (5) (for $N = 10^{19}$ cm^{-3} and a pump intensity of 10^{13} W/cm^2 we obtained $\nu_1/c = 0.83$ in He). This effect is also explained by the atoms parked at the upper level, thus significantly reducing the number of atoms available for photoionization. The photoionization time, η_{He}, which is also the width of the IFSR soliton, becomes shorter, $\eta_{\text{He}} \approx (1 - \alpha_{\text{He}})\eta_0$.[11]

Figure 2 shows a numerically obtained steady-state solution for the IFSR soliton in Li ($\sigma_2^p = \sigma_1^s = 0$). Here again we see a significant inhibition of the photoionization process (for a particular SERS in Li, $\alpha_{\text{Li}} \approx 0.83$), but the shape of the IFSR pulse is drastically different. According to Eqs. (2), in an ideal stationary solution (i.e., that resulting from infinitely long propagation) we have two semi-infinite pulses (one pumping and one Stokes), with the leading edge of the pump pulse (the photoionization front) coinciding with the trailing edge of the Stokes pulse. In practice, although the trailing edge of the Stokes pulse travels with the same velocity as the photoionization front, its leading edge travels much faster (actually with the velocity of light in neutral media). The length of such an almost rectangular pulse thus increases linearly with the distance traveled in media.[5] Therefore in the x-ray IFSR soliton in Li we have a strong Stokes precursor, arriving at the end of the cell significantly ahead of pumping. This effect can be used for measurements as well as for

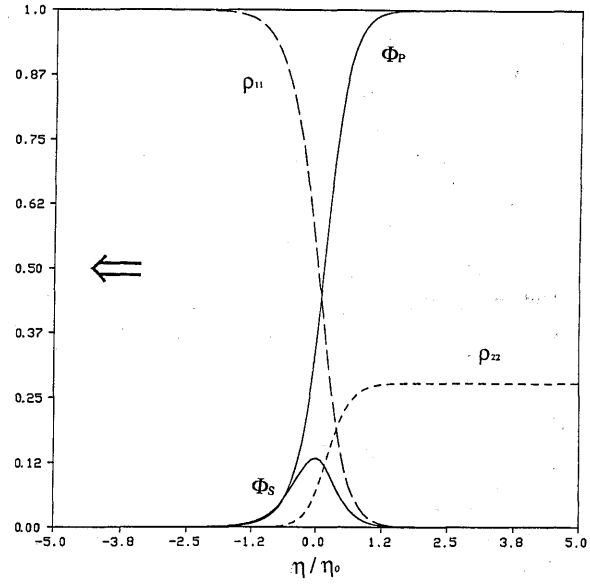

Fig. 1. Pump Φ_p and Stokes Φ_s photon fluxes [normalized to $\Phi_p(\infty) = \Phi_0$] and the population of the ground ρ_{11} and excited ρ_{22} levels as a function of the normalized retarded time η/η_0 for an IFSR soliton in He (for η_0 see the text). The arrow indicates the direction of pulse propagation.

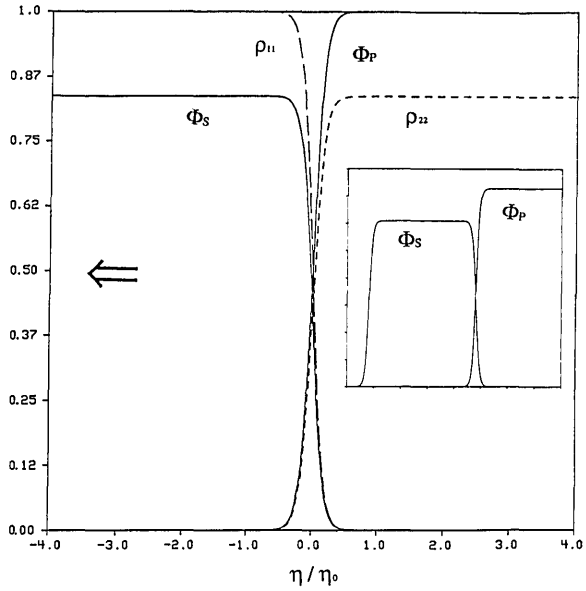

Fig. 2. Same as in Fig. 1, for an IFSR soliton in Li. Inset: the pump and Stokes pulses in a finite cell in Li.

a pilot warning of the trailing photoionization front. For a propagating distance of 10 cm, a pump intensity of 10^{12} W/cm^2, and $N = 10^{18}$ cm^{-3} ($\sim$0.1 atm at $T = 800$ K), the warning time is $\sim$100 ps. We again obtain a useful relation by considering the constant of motion (with $\sigma_2^p = \sigma_1^s = 0$):

$$N\rho_{11} - \delta\Phi_p = \text{const.}, \qquad N\rho_{22} - \delta\Phi_s = \text{const.} \quad (7)$$

With the boundary conditions $\rho_{11}(-\infty) = 1$, $\Phi_p(-\infty) = 0$ and $\rho_{11}(\infty) = 0$, $\Phi_p(\infty) = \Phi_0$, we obtain $\delta = -N/\Phi_0$, i.e., $\nu_2 = \nu_{\text{ion}}$, where ν_{ion} is given by Eq. (5). Thus the photoionization-front velocity in x-ray SERS in Li is the same as in the absence of SERS. Using the boundary conditions $\rho_{22}(-\infty) = 0$, $\rho_{22}(\infty) = \alpha_{\text{Li}}$, and $\Phi_s(\infty) = 0$, we obtain the following simple relation among the degree of photoionization inhibition, α_{Li}, the photon flux of the Stokes soliton $\Phi_s(-\infty)$, and the pumping flux Φ_0:

$$\Phi_s(-\infty)/\Phi_0 = \alpha_{\text{Li}}. \quad (8)$$

Here the photoionization time is also shorter than η_0 and is given as $\eta_{\text{Li}} \approx (1 - \alpha_{\text{Li}})\eta_0$.[11]

In conclusion, we have predicted two major types of IFSR solitons that result from resonantly enhanced x-ray SERS. We have shown that in He the solitonlike Stokes pulse can carry significant energy, inhibit the photoionization, and increase its front velocity. In Li, the leading edge (precursor) of the Stokes pulse is moving much faster than the trailing (photoionization) edge; this IFSR soliton also inhibits photoionization.

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10. Note that we simplified the Maxwell–Bloch equations here by neglecting the nonlinear Stark shift Ω_{St} (which is justified in the typical case in which $\Omega_{\text{St}} \ll |\omega_{\text{DEL}} - \omega_L|$, where ω_{DEL} is the frequency of the double-excited intermediate level transition) and the corrections to the group velocities $D_{p,s}$ (see details in Ref. 5) and thus assuming that $n_{p,s} \approx 1$, which is justified in the x-ray domain. The behavior of IFSR solitons in the presence of an anti-Stokes signal, as well as the influence of the Stark shift, will be treated in future studies.
11. More accurately, $(\eta_{\text{IFSR}})^{-1} = [(1 - \alpha)\eta_0]^{-1} + (\eta_R)^{-1}$, where η_R is related to the SERS process only and is independent of the photoionization cross sections.