

# X-ray-stimulated electronic Raman scattering in neutral gases and inhibited ionization

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We theoretically demonstrate the feasibility of resonantly enhanced, stimulated electronic Raman scattering (SERS) of Se x-ray laser radiation in atomic He and Li and estimate the Stokes output. We show that, despite strong photoionization, the conversion efficiency of the Raman process can be considerably high. We also found that coherent SERS can significantly inhibit x-ray-induced photoionization.

## INTRODUCTION

Rapid developments in x-ray lasers make research on x-ray nonlinear optics both interesting and timely. Plasmas were the first to be considered x-ray nonlinear media.<sup>1</sup> Nonionized x-ray nonlinear media, however, may have a number of obvious advantages over plasma in making the experimental observation of the proposed processes much easier. In this paper (our preliminary results were published in Ref. 2) we study stimulated electronic Raman scattering (SERS) by an x-ray laser in (initially) nonionized atomic gases, which, like SERS in the visible and infrared domain,<sup>3</sup> may serve as an important tool in obtaining new bright coherent sources in the x-ray domain. X-ray Raman scattering was studied previously<sup>4</sup>; however, those studies dealt primarily with hard-x-ray (~8 keV) spontaneous Raman scattering in crystals.

Realistically, x-ray SERS can be observed only under resonant enhancement; i.e., the pump frequency should be close to some atomic transition frequency. X-ray laser photon energy (~50–300 eV), however, is much larger than the binding energy of optical electrons in any neutral atom, so that no resonances with optical transitions are possible. Instead we suggest using so-called core-excited, or multiple-excited, atomic states with at least one of the inner electrons excited.<sup>5</sup> In particular, we found that some double-excited levels of He and Li atoms are resonant to the radiation of the Se<sup>24+</sup> x-ray laser developed at Lawrence Livermore National Laboratory.<sup>6</sup> The most promising for resonantly enhanced SERS appear to be transitions resonant to the 209.78-nm (59.10-eV) line of the Se x-ray laser; these transitions are presented in Fig. 1 with the necessary atomic information. The energy of He and Li double-excited levels are given in Refs. 7 and 8 and in 9, respectively; absorption oscillator strengths  $f_{13}$  and  $f_{23}$  are calculated by us from transition probabilities given in Ref. 7;  $f_{13}$  for Li is given in Ref. 10, whereas  $f_{23}$  is assumed by us to be equal to the respective value of Li-like B III calculated from the transition probability given in Ref. 11. Finally, the energy of the single-excited levels is taken from Ref. 12. The expected Stokes wavelengths are 32.2 and 22.3 nm in He and Li, respectively. The small-signal cw exponential

gain in the situation of resonant enhancement, in both cases, can be calculated<sup>3,13</sup> as  $G = I_p N g = I_p N \hbar \omega_s K^2 / 2\Gamma$ , with

$$K^2 = \frac{\pi^2 r_e^2 c^2}{\hbar^2} \frac{f_{13} f_{23}}{\Omega_{31} \Omega_{32}} \frac{1}{(\Omega_{31} - \omega_p)^2}, \quad (1)$$

where  $I_p$  is the pump intensity,  $N$  is the atomic-number density,  $g$  is the gain factor,  $r_e$  is the classical electron radius,  $\Gamma$  is the width of the Raman transition,  $\omega_s$  ( $\omega_p$ ) is the Stokes (pump) frequency,  $\Omega_{31}$  ( $\Omega_{32}$ ) is the frequency difference between the intermediate and the ground (final) state of the atom, and  $f_{13}$  ( $f_{23}$ ) is the oscillator strength of the transition from the ground (final) to the intermediate state. For  $\Gamma = \Gamma_D$  (Doppler broadening) and  $N$  in units of  $10^{18} \text{ cm}^{-3}$ , we obtain for He  $g_{\text{He}} = 2 \times 10^{-12} \text{ W}^{-1} \text{ cm}$  and for Li  $g_{\text{Li}} = 1.5 \times 10^{-11} \text{ W}^{-1} \text{ cm}$ . Stokes-output buildup competes with absorption, which in the soft-x-ray domain is largely due to photoionization. In He direct photoionization dominates, with cross sections for the Stokes and the pumping radiation being  $\sigma_s \approx \sigma_p \approx 10^{-18} \text{ cm}^2$ .<sup>14</sup> In Li direct photoionization is negligible compared with the absorption to the intermediate autoionizing double-excited level. The cross sections of the latter can be calculated as<sup>2</sup>

$$\begin{aligned} \sigma_p &= 2\pi r_e c f_{13} \Gamma_3 (\Omega_{31} - \omega_p)^{-2}, \\ \sigma_s &= 2\pi r_e c f_{23} \Gamma_3 (\Omega_{32} - \omega_s)^{-2} \end{aligned} \quad (2)$$

for the pump and the Stokes radiation, respectively. Here  $\Gamma_3 = 2.6 \text{ meV}$  (Ref. 9) is the autoionization width of the intermediate level. Using the pertinent atomic data (see Fig. 1), we obtain  $\sigma_p = 7 \times 10^{-19} \text{ cm}^2$  and  $\sigma_s = 4 \times 10^{-21} \text{ cm}^2$ . Thus, the characteristic absorption length is  $(N\sigma_p)^{-1} \approx 1 \text{ cm}$  for  $N = 10^{18} \text{ cm}^{-3}$ . Comparing this absorption length with the small-signal gain length, we find that to obtain a significant Stokes output would require pump intensities of the order of  $10^{12} - 10^{14} \text{ W/cm}^2$ . Such intense radiation would, however, fully ionize the medium in a much shorter time (e.g., ~1 ps for an intensity of  $10^{13} \text{ W/cm}^2$ ) than the typical pulse duration of the Se x-ray laser (~100 ps). Thus effective Stokes generation can take place only at the leading edge of the laser pulse, before the full photoionization sets in. Since

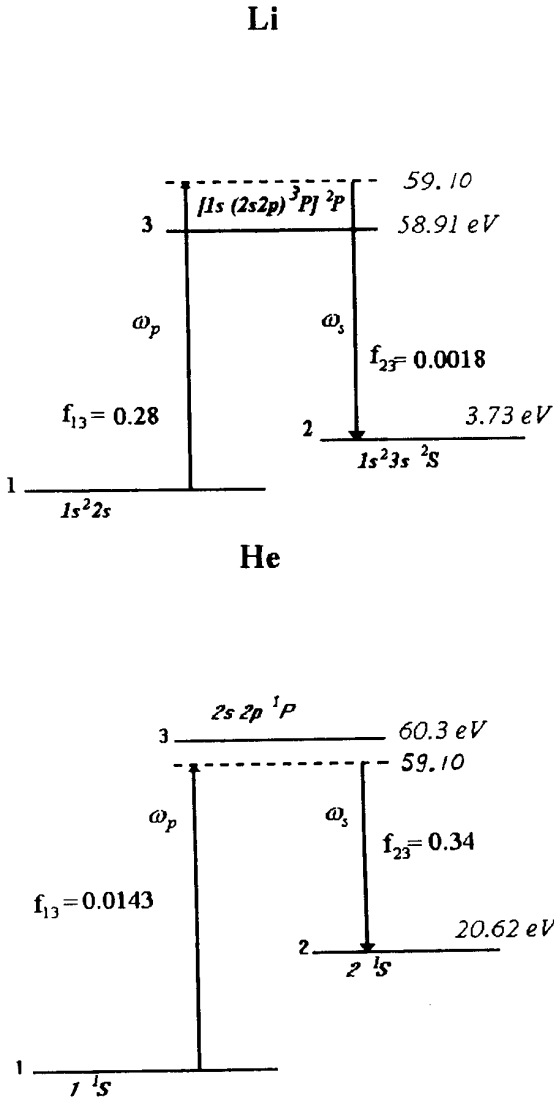


Fig. 1. Essential-level diagrams for suggested x-ray SERS in Li and He.

the excited Raman levels in He and Li have lifetimes of a few microseconds<sup>15</sup> and nanoseconds,<sup>16</sup> respectively, this effective Stokes time,  $\tau_{\text{ion}}$ , is shorter than both the inhomogeneous and the homogeneous broadening times, so that the x-ray SERS under consideration is a coherent process.<sup>17-20</sup>

It should be noted that the neutral atoms can be ionized also by the free electrons produced in the photoionization process. The cross sections of collisional (electron-impact) ionization of the ground-level He (Li) atoms for  $\sim 35$ -eV ( $\sim 54$ -eV) electrons produced in photoionization are  $\sim 5 \times 10^{-18} \text{ cm}^2$  ( $\sim 2 \times 10^{-16} \text{ cm}^2$ ).<sup>21</sup> Assuming a photoelectron density of  $\sim 5 \times 10^{17} \text{ cm}^{-3}$ , we obtain the He (Li) atomic lifetime that is due to direct (i.e., before the photoelectrons become thermalized) collisional ionization of  $\sim 1 \text{ ns}$  ( $\sim 20 \text{ ps}$ ). Thus, since these lifetimes are much longer than the Stokes time related to photoionization,  $\tau_{\text{ion}} \sim 1 \text{ ps}$ , electron-impact ionization will be neglected here. The free electrons can also ionize atoms from the excited Raman level. However, since each atom

is transferred to that level only after Raman scattering takes place, this process has a relatively small influence on the generation of Stokes radiation. In any case, we estimate, following Ref. 22, that the lifetimes of the excited levels are 10 (2) times longer than the Stokes time for He (Li) atoms (for  $I_p = 10^{13} \text{ W/cm}^2$  and  $N = 10^{18} \text{ cm}^{-3}$ ), and therefore we neglect collisional ionization of the excited level as well.

## MAXWELL-BLOCH ANALYSIS

To the best of our knowledge, coherent SERS in the presence of strong absorption and the associated atomic depletion has never been considered. In general, such a process is governed by coupled Maxwell-Bloch equations,<sup>18,19</sup> which in our case have to be modified to include the effect of photoionization. We assume plane-wave pulses and represent the pump ( $E_p$ ) and the Stokes ( $E_s$ ) fields as

$$E_{p,s} = \text{Re}\{E_{p,s} \exp[i(\omega_{p,s}t - k_{p,s}z)]\},$$

where  $k_{p,s} = n_{p,s}\omega_{p,s}/c$  and  $E_{p,s}$  are slowly varying, complex field envelopes. The photoionization is described by field-dependent decay terms proportional to the linear combination of the intensities  $|E_p|^2$  and  $|E_s|^2$ , with the respective coefficients  $\beta_j^p$  and  $\beta_j^s$  ( $j = 1, 2$ ) ascribed to each level  $j$ . These coefficients depend on the photoionization cross sections  $\sigma_p$  and  $\sigma_s$ , respectively (see below). Introducing field-dependent relaxation rates  $w_j = \beta_j^p|E_p|^2 + \beta_j^s|E_s|^2$  and the retarded coordinate system  $\tau = t - z/c$ ,  $\xi = z$ , one can write dynamic equations for the field envelopes  $E_p$  and  $E_s$ , the populations  $\rho_{11}$  and  $\rho_{22}$ , and the slowly varying envelope  $\rho$  of the nondiagonal elements of the density matrix, defined as  $\rho_{21} = \rho_{12}^* = i\rho \exp[i(k_p - k_s)z - i\omega_0t]$ :

$$\partial \rho_{jj} / \partial \tau = (-1)^j K \text{Re}(\rho E_p E_s^*) - w_j \rho_{jj} \quad (j = 1, 2), \quad (3a)$$

$$\begin{aligned} \partial \rho / \partial \tau = & -i\Omega_{\text{St}}\rho + (1/2)(\rho_{11} - \rho_{22})KE_p^*E_s \\ & - (1/2)(w_1 + w_2)\rho, \end{aligned} \quad (3b)$$

$$\begin{aligned} \partial E_p / \partial \xi = & (N\hbar\omega_p/\epsilon_0c)[- \rho KE_s \\ & - E_p(iD_p + \beta_1^p\rho_{11} + \beta_2^p\rho_{22})], \end{aligned} \quad (3c)$$

$$\begin{aligned} \partial E_s / \partial \xi = & (N\hbar\omega_s/\epsilon_0c)[ \rho KE_p \\ & - E_s(iD_s + \beta_1^s\rho_{11} + \beta_2^s\rho_{22})], \end{aligned} \quad (3d)$$

where  $\Omega_{\text{St}}$  is a nonlinear Stark shift,<sup>23</sup>

$$\begin{aligned} \Omega_{\text{St}} = & (4\hbar)^{-1}\{[\chi_1(\omega_p) - \chi_2(\omega_p)]|E_p|^2 \\ & + [\chi_1(\omega_s) - \chi_2(\omega_s)]|E_s|^2\}, \end{aligned} \quad (4)$$

$D_{p,s}$  are corrections to the group velocities at the respective pump and Stokes frequencies,

$$D_{p,s} = (2\hbar)^{-1}[\chi_1(\omega_{p,s})\rho_{11} + \chi_2(\omega_{p,s})\rho_{22}], \quad (5)$$

$\epsilon$  is the vacuum permittivity, and  $\chi_1$  ( $\chi_2$ ) is the linear susceptibility of the ground (excited) level (note that in

the case of a purely two-level system  $\chi_1 = \chi_2$ ). In He,  $\beta_2^p = \beta_2^s \approx 0$  and  $\beta_{p,s}^1 = \sigma_{p,s} \epsilon_0 c / 2\hbar \omega_{p,s}$ , where  $\sigma_{p,s} \approx 10^{-18} \text{ cm}^2$ . In Li,  $\beta_1^s = \beta_2^p \approx 0$ ,  $\beta_2^s \approx \sigma_s \epsilon_0 c / 2\hbar \omega_s$ , and  $\beta_1^p \approx \sigma_p \epsilon_0 c / 2\hbar \omega_p$ , where  $\sigma_{p,s}$  are given by Eqs. (2).

Equations (3) are derived from the full three-level set of Bloch equations (see, e.g., Ref. 24) by introduction of fast relaxation for the population  $\rho_{33}$  of the intermediate level and the off-diagonal elements  $\rho_{13}$  and  $\rho_{23}$ . Then, by assuming  $\rho_{33} \approx \text{constant} = 0$  and  $\Gamma_3 \rho_{j3} \gg \partial \rho_{j3} / \partial \tau$  (where  $j = 1, 2$  and  $\Gamma_3$  is the linewidth of the intermediate level), one can solve these equations exactly for  $\rho_{j3}$ , resulting in Eqs. (3a)–(3b).<sup>23</sup>

For further calculations we simplify Eqs. (3) by neglecting the linear susceptibilities of atoms in the x-ray domain,  $\chi_1(\omega_{p,s}) \approx \chi_2(\omega_{p,s}) \approx 0$ , which allows us to neglect the Stark shift  $\Omega_{St}$  and the corrections to the group velocities  $D_{p,s}$  in Eqs. (3)<sup>25,26</sup> (and thus to assume that  $n_{p,s} \approx 1$ ). Under these conditions there is no phase modulation, and Eqs. (3) can be expressed in terms of the intensities  $I_{p,s} = |E_{p,s}|^2$  and the envelope  $\rho$ , which now becomes purely real:

$$\partial \rho_{jj} / \partial \tau = (-1)^j U - w_j \rho_{jj} \quad (j = 1, 2), \quad (6a)$$

$$\partial \rho / \partial \tau = U(\rho_{11} - \rho_{22}) / 2 - (w_1 + w_2) \rho / 2, \quad (6b)$$

$$\partial I_i / \partial \xi = (2N\hbar\omega_i / \epsilon_0 c) [S_i U \rho - (\beta_1^i \rho_{11} + \beta_2^i \rho_{22}) I_i] \quad (i = p, s), \quad (6c)$$

where  $S_p = -S_s = -1$  and  $U^2 = K^2 I_p I_s$ .

Because of the autoionization, Eqs. (6) support conservation of the number of neither atoms ( $\rho_{11} + \rho_{22} \neq 1$ ) nor photons (i.e., the Manley–Rowe relationship does not hold). In their simple form, with  $\beta_{1,2}^{s,p} = 0$ , Eqs. (6) have been the subject of numerous studies<sup>17–20</sup>; it is believed that, in general, no analytical solution for them exists. Some approximations, however, permit an analytical description of the process. For instance, the assumptions  $\rho_{11} = 1$ ,  $\rho_{22} = 0$ , and  $I_p(0, \tau) = I_p(z, \tau)$  leave only two equations, which can be solved analytically.<sup>17</sup> In the small-area approximation,<sup>18</sup> the solution is further simplified, displaying an exponential growth of the Stokes energy, with the gain coefficient proportional to the total energy of the pump pulse. It is well known from numerical studies<sup>19</sup> that, without the above-mentioned approximations, the exponential growth eventually saturates because of either atomic depletion or pump depletion. Atomic depletion, that is, the depopulation of the ground level ( $\rho_{11} \ll 1$ ), results in the linear ( $\sim N\hbar\omega_s z$ ) growth of the Stokes energy. Pump depletion represents an ultimate limitation to the SERS process: at a sufficiently large propagation distance, the Stokes energy growth may saturate because of the total conversion of the pump energy to the energy of the Stokes pulse.

If the relaxation times  $(w_j)^{-1}$  are much longer than the pulse duration  $\tau_p$ , one can neglect them in Eqs. (6a)–(6b) and use the existing approximate analytical solutions. For example, the exponential approximation with only  $\beta_1^p \neq 0$  results in an exponential growth, saturating after the characteristic absorption length of the pump  $(N\sigma_p)^{-1}$ . In this paper, however, we investigate the case in which relaxation times that are due to photoionization cannot be neglected. Equations (6) show that, if we assume that  $I_p(0, \tau) \gg I_s(0, \tau)$  and  $\rho_{22} = 0$  for  $\tau < 0$ , then, at least

in the early stage of the process, the most significant contribution to the Raman process comes from the terms proportional to  $\beta_1^p$  in Eqs. (6a) and (6b) and from  $\beta_1^p$  and  $\beta_1^s$  in Eq. (6c).

## EXPONENTIAL SERS GAIN: NUMERICAL RESULTS AND ANALYTICAL APPROXIMATIONS

Figure 2, curves 1 and 3, show typical results of a numerical solution of Eqs. (6) for Li and He, respectively. The normalized Stokes-pulse energy flux  $J_s(z)/J_p$  is shown as a function of the cell length  $z$  for  $N_{Li} = 10^{18} \text{ cm}^{-3}$  [corresponding to a pressure of  $\sim 0.1 \text{ atm}$  (76.0 Torr) and  $T = 800 \text{ K}$ ],  $N_{He} = 2 \times 10^{18} \text{ cm}^{-3}$  [ $\sim 0.08 \text{ atm}$  (60.8 Torr) at  $T = 300 \text{ K}$ ], the initial pump-pulse energy flux  $J_p = 10^3 \text{ J/cm}^2$  in He and  $J_p = 2.3 \times 10^2 \text{ J/cm}^2$  in Li, and the initial normalized energy  $J_s(0)/J_p = 10^{-12}$ . In our calculations we assumed that the seeded Stokes pulse has the same (i.e., Gaussian) shape and duration as the pump pulse.

Figure 2 clearly shows two distinct stages of this Raman process, namely, exponential growth and saturation. First we concentrate on the exponential region. Figures 3(a) and 4(a) show  $I_p(z, \tau)$  for different cell lengths  $z$  in Li and He, respectively. One can see that, owing to photoionization, the leading edge of the pump pulse is being sharpened and the pulse duration is shortening. In the retarded coordinate system this is seen as the retreating of the leading edge of the pulse toward larger  $\tau$ . Figure 3(b) shows  $I_s(z, \tau)$  for various values of  $z$  inside the exponential region for Li (the picture is qualitatively the same in He). Apart from some short initial stage, the width of the Stokes pulse remains constant, and the peak of the intensity follows the retreating leading edge of the pump. The most important result here is that the exponential growth region

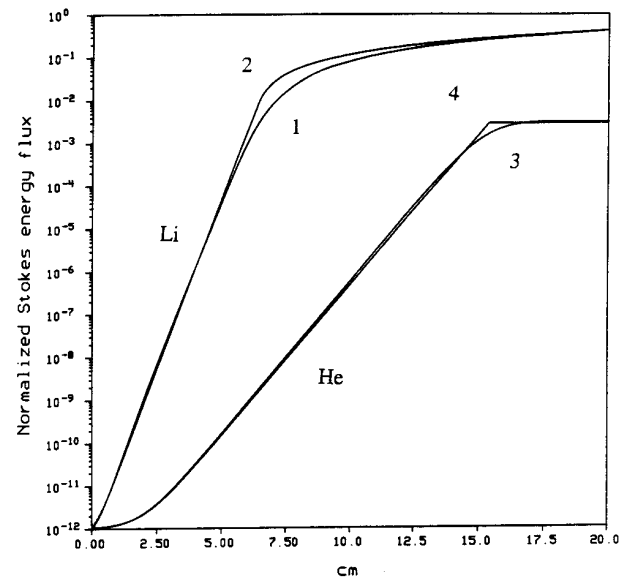
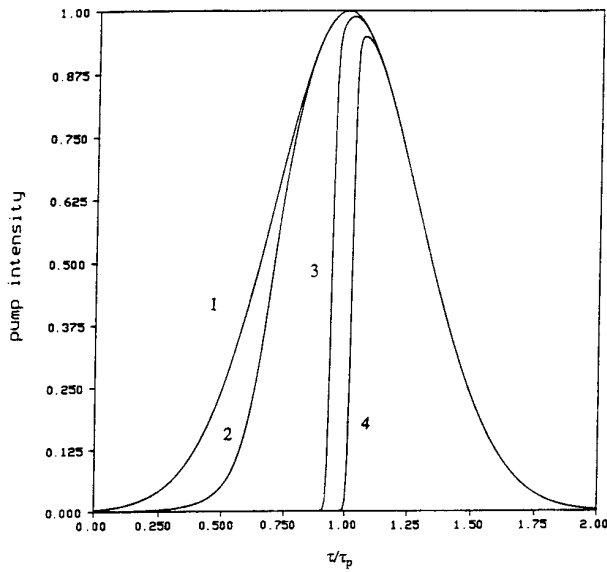
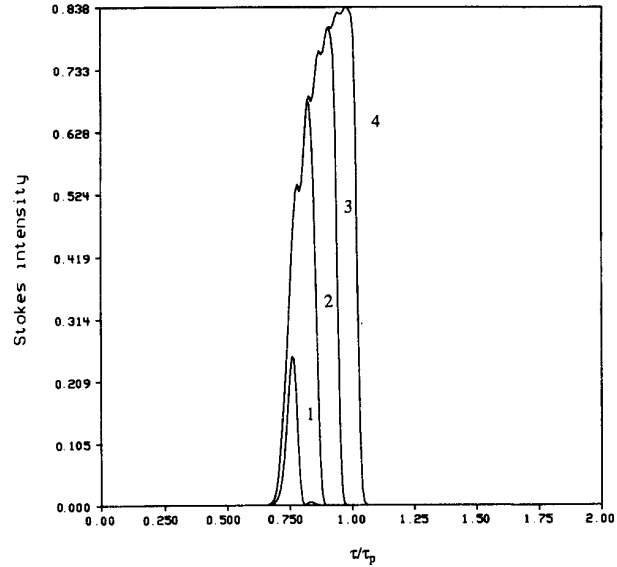


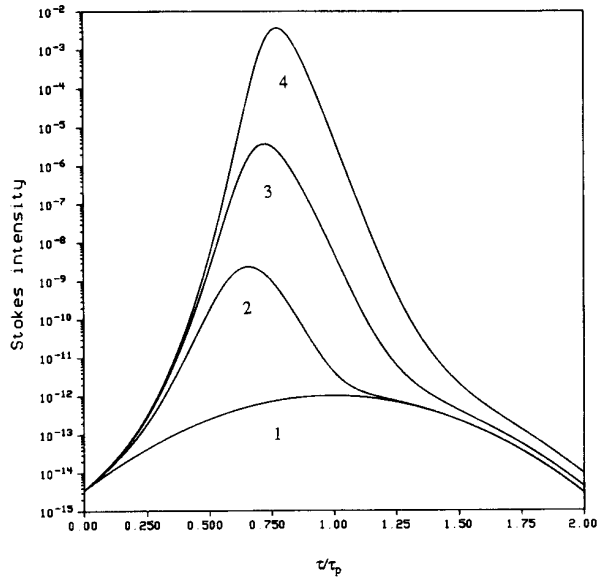
Fig. 2. Normalized Stokes energy flux  $J_s(z)/J_p$  as a function of the cell length  $z$  for  $N_{He} = 2 \times 10^{18} \text{ cm}^{-3}$ ,  $N_{Li} = 10^{18} \text{ cm}^{-3}$ , and  $J_s(0)/J_p = 10^{-12}$ . Curves 1 and 3, numerical results; curves 2 and 4, approximate analytical model.



(a)



(c)



(b)

Fig. 3. Normalized intensity of (a) pump,  $I_p(z, \tau)/I_{p\max}$ , and (b), (c) Stokes,  $I_s(z, \tau)/I_{p\max}$ , pulses in Li versus normalized retarded time  $\tau/\tau_p$ . (a) Curves 1,  $z = 0$  cm; 2,  $z = 4$  cm; 3,  $z = 16$  cm; 4,  $z = 20$  cm. (b) Curves 1,  $z = 0$  cm; 2,  $z = 2$  cm; 3,  $z = 4$  cm; 4,  $z = 6$  cm. (c) Curves 1,  $z = 8$  cm; 2,  $z = 12$  cm; 3,  $z = 16$  cm; 4,  $z = 20$  cm.

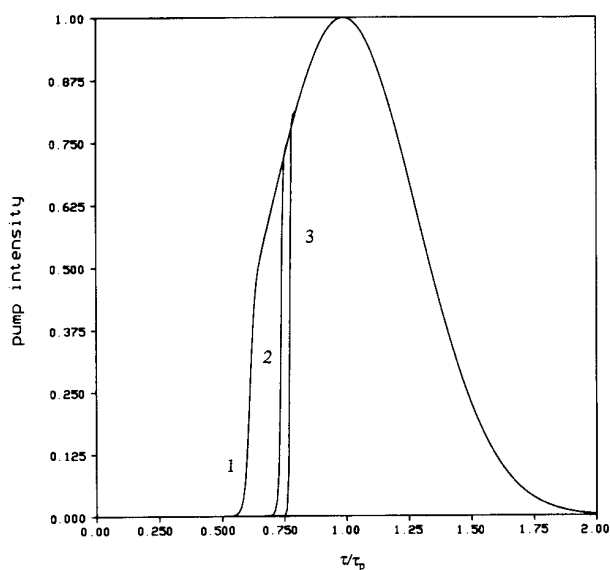
is much longer than the characteristic absorption length of the pump  $(N\sigma_p)^{-1}$ , which is 1.5 cm for Li and 0.5 cm in He for conditions in Fig. 2. This seemingly surprising result can be qualitatively explained as follows. Inside the almost constant pulse width [see Fig. 3(b)] the Stokes intensity grows exponentially toward the trailing edge of the pulse, where it falls rapidly owing to the photoionization of the media. (This picture is consistent with the previous reports on the shape of the Stokes pulses in the transient Raman scattering.<sup>17</sup>) Thus, since the Stokes energy is concentrated at the trailing edge of the Stokes pulse, the losses caused by the retreat of the leading edge of the pump are negligible compared with the exponential amplification of the trailing edge of the Stokes pulse. These results suggest that an approximate solution may be obtained by neglecting the terms proportional to  $\beta$

parameters in Eqs. (6), except for  $\beta_1^s$  in Eq. (6c), and by restricting the solution to  $0 < \tau < \tau_{\text{ion}}$ , where  $\tau_{\text{ion}}$  is determined by the photoionization rate by means of

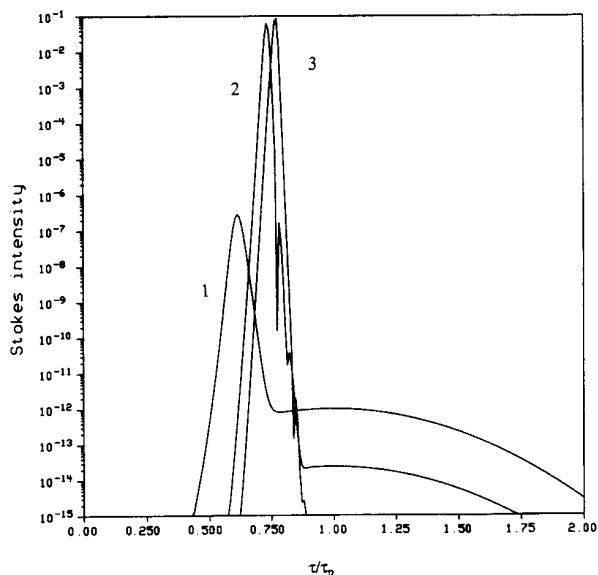
$$\int_0^{\tau_{\text{ion}}} \beta_1^p E_p^2(0, \tau) d\tau = \gamma, \quad (7)$$

where  $\gamma = \text{constant} = O(1)$ ; as we find below,  $\gamma = 0.77$ . The resulting equations are now similar to the equations discussed in Refs. 17–20. In the small-area approximation the Stokes energy flux is expected to exhibit exponential growth, with the gain coefficient being proportional to the pump energy flux contained in the interval  $0 < \tau < \tau_{\text{ion}}$ . This energy flux,  $J_p^{\text{eff}}$ , can be calculated from Eq. (7):

$$J_p^{\text{eff}} = \gamma \epsilon_0 c (2\beta_1^p)^{-1} = \gamma \hbar \omega_p / \sigma_p = N \hbar \omega_p l_p, \quad (8)$$



(a)



(b)

Fig. 4. Normalized intensity of (a) pump,  $I_p(z, \tau)/I_{p\max}$ , and (b) Stokes,  $I_s(z, \tau)/I_{s\max}$ , pulses in He versus normalized retarded time  $\tau/\tau_p$ . (a) Curves 1,  $z = 8$  cm; 2,  $z = 16$  cm; 3,  $z = 20$  cm. (b) Curves 1,  $z = 8$  cm; 2,  $z = 16$  cm; 3,  $z = 20$  cm.

where we introduce the effective pump absorption length  $l_p = \gamma(N\sigma_p)^{-1}$ . For Li, with small Stokes absorption neglected,  $\sigma_s \ll \sigma_p$  [for example,  $(N\sigma_s)^{-1} \approx 300$  cm for  $N = 10^{18}$  cm $^{-3}$ ], the Stokes exponential gain coefficient is

$$G = K^2(N^2\hbar^2\omega_s\omega_p/\epsilon_0^2c^2)l_p. \quad (9)$$

The best fit between Eq. (9) and the slope of curve 1 in Fig. 2 was obtained with  $\gamma = 0.77$ . Since the Raman process occurs only near the leading edge of the pump pulse, only a small portion of the Stokes pulse is exponentially amplified:

$$J_s(z) = J_s(0) + J_s^{\text{eff}} \exp(Gz), \quad (10)$$

where we introduced the effective initial Stokes energy flux  $J_s^{\text{eff}}$  instead of  $J_s(0)$ . As can be seen from Fig. 3(b), the energy of the Stokes pulse inside the effective pulse duration  $\tau_{\text{ion}}$  consists of two parts: amplified Stokes energy from the retreating leading edge and unamplified Stokes energy from the trailing edge of the pulse. Assuming, as in our numerical simulation [see Figs. 3(a) and 3(b)], that pump and Stokes pulses have the same shape and duration, the Stokes energy flux contained in the time interval  $0 < \tau < \tau_{\text{ion}}$  is  $\bar{J}_s = J_s(0)J_p^{\text{eff}}/J_p$ . For a large exponential gain,  $l_p \gg l_g = \gamma G^{-1}$ , as in Li, the Stokes input from the trailing edge can be neglected, so

$$J_s^{\text{eff}} \approx \gamma_1 J_s(0) N \hbar \omega_p l_p / J_p, \quad (11a)$$

where  $\gamma_1 = O(1)$  is a dimensionless correction factor; for Gaussian pulses,  $\gamma_1 = 4.8$  yields the best fit to the numerical results. In the opposite limit,  $l_p \ll l_g$ , the effective initial Stokes energy flux can be estimated as

$$J_s^{\text{eff}} \approx \gamma_2 J_s(0) N \hbar \omega_p l_g / J_p. \quad (11b)$$

The best fit to the numerical results in He was obtained from relation (11b) with  $\gamma_2 = 3.6$ . In He, Stokes absorption cannot be neglected, and the energy flux growth is thus given by

$$J_s(z) = J_s(0) + J_s^{\text{eff}} \exp[(G - 1/l_s)z], \quad (12)$$

where  $l_s = \gamma_s(N\sigma_s)^{-1}$ . The best fit to the numerical solution corresponds to  $\gamma_s \approx 2.5$ . This increase in the Stokes absorption length, compared with the absorption length without strong photoionization, occurs because the Stokes pulse is located inside the effective pump pulse, where the ground population is significantly reduced. It is easy to see from Eq. (12) that, in general, under the conditions discussed in this paper, i.e., for  $\tau_{\text{ion}} \ll \tau_p$ , amplification of the Stokes signal is possible only if  $l_s G > 1$ . Defining the characteristic Raman cross section as  $\sigma_R \equiv (G/N^2 l_p)^{1/2}$  and noticing that because of the definition of  $G$  [Eq. (9)]  $\sigma_R$  depends only on the spectroscopic parameter of an atom, we can express the condition  $l_s G > 1$  in a purely spectroscopic form:

$$\sigma_R^2 = K^2 \hbar^2 \omega_s \omega_p / \epsilon_0^2 c^2 > \sigma_p \sigma_s / \gamma \gamma_s. \quad (13)$$

For not very large pump energy, i.e., when  $\tau_p < (w_j)^{-1}$ , one can neglect the photoionization terms  $w_j$  in Eqs. (6a) and (6b), which results in almost the standard equations for coherent Raman scattering.<sup>18,19</sup> In the small-area approximation<sup>18</sup> we then expect the Stokes energy to exhibit exponential gain saturating at  $z \approx l_p$ . Suppose now that the pump energy is increased so that, while the strength of the Raman process also increases, photoionization intensifies and becomes faster than the pulse duration,  $\tau_p > (w_1)^{-1}$ . Will this lead to a significant increase or decrease in the Stokes output? In Li the condition  $\tau_p = (w_1)^{-1}$  is satisfied at the pump energy flux of  $\sim 9$  J/cm $^2$ . Such pumping yields an exponential gain of  $\sim 3.6$  at  $z = l_p$ , and, under the same conditions as in Fig. 2, the conversion efficiency is  $\sim 6 \times 10^{-9}$ . This is

in striking contrast to the situation depicted in Fig. 1 for  $\tau_p \gg \omega_1^{-1}$ , in which case the conversion efficiency approaches unity. It is therefore clear that an increase in the pump intensity to the point  $\tau_p \gg (\omega_1)^{-1}$  can result in an enormous increase not only in the absolute Stokes energy output but also in the conversion efficiency. In addition, as can be seen from Figs. 3(a), 3(b), and 4, it is possible, by operating in the exponential region, to obtain very short, high-energy Stokes pulses, as well as to sharpen the Se x-ray laser pulse. Both the Stokes pulse duration and the pump rise time are of the order of  $\tau_{\text{ion}}$ .

### SATURATION IN He AND Li: DIFFERENT PHOTOIONIZATION MECHANISMS

In the saturation region (see Fig. 2) there is a qualitative difference in the behavior of the Stokes output in Li and in He. In Li the exponential growth gradually becomes linear. At  $z \approx 8\text{--}10$  cm [see Fig. 3(c)] the intensity of the Stokes pulse becomes comparable with that of the pump. It is at this point that the analogy between this process and the one described by neglecting the  $\beta$  parameters and restricting the solution to  $\tau < \tau_{\text{ion}}$  in Eqs. (6) breaks down. In the latter process there can be no substantial additional amplification beyond this point, since almost all the energy of the pump has already been converted to the Stokes pulse. In the former process, however, there may still be a significant amount of energy in the still unused, trailing edge of the pump pulse. This energy is now being converted to the Stokes pulse, whose amplitude from now on remains almost constant and whose duration increases linearly with distance. The Stokes energy in the linear region can be described by the equation

$$J_s(z) = J_s(z_2) + \alpha N \hbar \omega_s (z - z_2), \quad (14)$$

where the excited level population after the passage of the pulse  $\alpha = \rho_{22}(z, \infty) = \text{constant}$  was numerically evaluated to be  $\alpha \approx 0.83$ . The distance  $z_2$  at which the pump depletion inside the effective pulse occurs [i.e.,  $J_s(z_2) \approx J_p^{\text{eff}}$ ] can be roughly estimated by

$$z_2 \approx \gamma_3 G^{-1} \ln(J_p^{\text{eff}}/J_s^{\text{eff}}) = \gamma_3 G^{-1} \ln(N \hbar \omega_p l_p / J_s^{\text{eff}}), \quad (15)$$

where  $\gamma_3 = O(1)$  is again a correction factor; the best numerical fit was found for  $\gamma_3 = 0.9$ . Another important characteristic length of the process, the distance  $z_1$  at which atomic depletion occurs, can be estimated by assuming that at  $z_1$  the Stokes photon flux should be roughly equal to half the number of atoms in the amplification length  $l_g$ ; then

$$z_1 \approx G^{-1} \ln(N \hbar \omega_s l_g / 2 J_s^{\text{eff}}). \quad (16)$$

Since in Li  $l_g < l_p$ , atomic depletion occurs before pump depletion:  $z_1 < z_2$ . Our numerical calculations show that  $\alpha \approx 0.83$ , independent of atomic-number density, pump or Stokes intensities, or cell length. This constant can also be estimated analytically. We notice first that in the absence of photoionization, i.e., if  $\beta_1^p = 0$ , every

atom in the linear region is transferred to the excited Raman level within a time period  $\tau_R \approx \pi(K E_p E_s)^{-1}$ . Since only the ground-state Li atoms can be ionized by either pumping or Stokes pulses, it is reasonable to estimate  $\alpha$  by calculating the number of atoms ionized in half the time  $\tau_R$ :

$$\alpha \approx \exp(-\beta_1^p E_p^2 / \tau_R / 2) = \exp(-\pi \beta_1^p / 2K), \quad (17)$$

where  $K$  is given by Eq. (1). In Li,  $\beta_1^p \approx \sigma_p \epsilon_0 c / 2 \hbar \omega_p$ , resulting in the estimate  $\alpha \approx 0.74$ , in good agreement with the numerical result. Figure 2 (curve 4) shows the results of the approximate analytical model for Li, with the assumption of exponential growth up to and including  $z_2$  and linear growth for  $z > z_2$ .

In He with  $N = 2 \times 10^{18} \text{ cm}^{-3}$  the exponential growth saturates completely at  $z \approx 17$  cm (with a maximum Stokes flux of  $\sim 3 \text{ J/cm}^2$ ), since the amplification of Stokes energy starts to decrease after the exponential region and eventually becomes equal to the absorption. From this point on, the number of Stokes photons generated per unit cell length is exactly balanced by the number of photons absorbed by the medium. Figure 4 shows the pump and the Stokes pulse intensities versus  $z$  in He. As one can see, there is no significant pump depletion, and the shape of the Stokes pulse is constant. Essentially these results predict [Fig. 2(b)] a solitonlike behavior of the Stokes component at the photoionization front of the x-ray radiation, which is related to the rate balance of SERS and photoionization. Now, if it is assumed that the saturation comes from the atomic depletion only, and that in the saturation region  $G \approx l_s^{-1}$ , the maximum Stokes flux can be estimated from the following balance equation:

$$(G + l_s^{-1}) J_{\text{max}} / \hbar \omega_s \approx 2 l_s^{-1} J_{\text{max}} / \hbar \omega_s \approx N/2. \quad (18)$$

The left-hand side of relation (18) is an approximate number of atoms per unit volume removed from the ground level by Stokes amplification and absorption. The critical cell length  $z_{\text{cr}}$  at which  $J_{\text{max}}$  energy flux is first reached may be evaluated with the assumption that exponential gain holds up to and including  $z_{\text{cr}}$ :

$$z_{\text{cr}} \approx \gamma_{\text{cr}} (G - 1/l_s)^{-1} \ln(J_{\text{max}}/J_s^{\text{eff}}). \quad (19)$$

The excited level population after the passage of the pulse in He was calculated numerically to be  $\alpha \approx 0.3$ . Figure 2 (curve 4) shows the results of the approximate analytical model as compared with the numerical solution for He, where we assumed an exponential region [Eq. (10)] up to and including  $z_{\text{cr}}$  ( $\gamma_{\text{cr}} = 1$ ) and total saturation ( $J_s = J_{\text{max}}$ ) after that. In the saturation region the x-ray Raman scattering described here results in a substantial transfer of the atomic population to excited Raman levels (83% of the atoms in Li and 30% in He). It is thus possible that the resulting population inversion may be used to observe x-ray-stimulated anti-Stokes scattering.

## SERS-INDUCED INHIBITION OF PHOTOIONIZATION AND SOLITONLIKE BEHAVIOR

Summarizing the results obtained so far, we can give a general description of the entire picture of SERS in the presence of strong pump-induced photoionization of the initially neutral gas. At the first stage, when the Stokes component is small, SERS grows exponentially with distance in both Li and He alike. At this stage the spatial dynamics of the pump itself is determined purely by the photoionization whose front travels with the velocity  $v_{\text{ion}}$ , readily found from Eqs. (6) as

$$v_{\text{ion}} = (1/c + N/\Phi_0)^{-1}, \quad (20)$$

where  $\Phi_0 = \Phi_p(\infty)$ ,  $\Phi_{p(s)}(\eta) = I_{p(s)}(\eta)\epsilon_0 c/2\hbar\omega_{p(s)}$  is the photon flux at the pump (Stokes) frequency, and  $\eta = t - z/v_{\text{ion}}$ , with the front spatial length  $\eta_0 = 2/\sigma_p\Phi_0$ . Equation (20) is obtained from Eqs. (6) under the conditions that  $I_s \ll I_p$  and that at  $\eta \rightarrow \infty$ ,  $\Phi_p \rightarrow \text{constant} = \Phi_0$  (i.e., the pump is a semi-infinite wave), which result in the photoionization front of the form  $I_p = (I_0/2)[\tanh(\eta/\eta_0) + 1]$ . At the second stage, when the Stokes component becomes relatively strong and saturation sets in, the two gases He and Li demonstrate different behavior.

In He, in which an atom can be efficiently ionized (by either Stokes or pump components) from the ground level only and photoionization from the excited level is negligible, the saturation mode can be described by a stable solitonlike Stokes pulse traveling with exactly the same velocity as the photoionization front and the leading edge of the pumping, which leaves in its wake He ions and excited neutral He atoms in the proportion  $(1 - \alpha)/\alpha$  (in the particular case considered above,  $\alpha = 0.3$  for He). Amazingly enough, the coherent interaction, i.e., SERS, is able to inhibit significantly the photoionization depletion of neutral atoms, with the degree of this inhibition being measured by  $\alpha$ . Related to this inhibited photoionization depletion there is also inhibited retardation of the photoionization front (this is true only in the case of He). Our calculations based on Eqs. (6) show that the photoionization front (and the leading edge of the pumping field) now travels much faster, such that

$$v_{\text{ion}}^{\text{He}} = [1/c + (1 - \alpha)N/\Phi_0]^{-1}. \quad (21)$$

Equation (21) is obtained from Eqs. (6) with the assumptions that the pump is again a semi-infinite wave, i.e.,  $\Phi_p(-\infty) = \Phi_s(-\infty) = \Phi_s(\infty) = 0$ ,  $\Phi_p(\infty) = \text{constant} = \Phi_0$ , and  $\rho_{11}(-\infty) = 1$ , resulting in the relation between  $\rho_{\Sigma} \equiv \rho_{11} + \rho_{22}$ ,  $\Phi_{\Sigma} \equiv \Phi_p + \Phi_s$ , and  $\delta^{\text{He}} \equiv 1/v_{\text{ion}}^{\text{He}} - 1/c$ :

$$\rho_{\Sigma} + \delta^{\text{He}}\Phi_{\Sigma}/N = 1. \quad (22)$$

Assuming that  $\rho_{\Sigma}(\infty) = \rho_{22}(\infty) = \alpha$ , one obtains Eq. (21). The result for  $v_{\text{ion}}^{\text{He}}$ , Eq. (21), was corroborated by our computer simulation with good accuracy.

In Li, owing to the near resonance of both pump and Stokes frequencies to the intermediate level, photoionization through that level can be caused only by the pump from the ground level or by the Stokes pulse from the excited level. The saturation mode (in the ideal situation) is again described by a semi-infinite pump wave whose

leading edge travels along with the photoionization front. It can readily be proved from Eqs. (6) that in this case the velocity of the photoionization front,  $v_{\text{ion}}$ , remains unchanged, i.e., is given by Eq. (20). The Stokes component in Li is a pulse with a flat top, whose trailing edge coincides with the leading edge of the pump and travels with the same velocity,  $v_{\text{ion}}$ . Its leading edge travels much faster, with the group velocity of light in the neutral gas (i.e., almost  $c$ ), running farther from the trailing edge while its top intensity remains almost constant. Therefore in Li vapor we expect a strong Stokes precursor to arrive at the end of the cell significantly ahead of the pump. This precursor can be used for measurements, as well as for warning of the coming photoionization front. The total energy of the Stokes component increases linearly with time (or distance) as  $J_s \sim t(c - v_{\text{ion}})$ . After sufficiently long propagation, the entire picture looks even simpler: the wave structure consists of two semi-infinite waves, such that the leading edge of one of them (the pump with the intensity  $\Phi_p(\infty) = \Phi_0$ ) coincides with the trailing edge of another [i.e., the Stokes precursor with  $\Phi_s(-\infty) = \Phi_{s\infty} < \Phi_0$ ]. From Eqs. (6) it can readily be shown that in this case the photon fluxes at each frequency are directly related to the respective populations:  $\Phi_p = \Phi_0(1 - \rho_{11})$  and  $\Phi_s = \Phi_{s\infty} - \rho_{22}\Phi_0$ . Hence, since  $\rho_{22}(\infty) = \alpha$ , we obtain the relationship between photon flux in the Stokes precursor  $\Phi_{s\infty}$  and the population  $\alpha$  of the excited yet neutral atoms in the wake of a photoionization front:

$$\Phi_{s\infty} = \alpha\Phi_0. \quad (23)$$

In our computer simulation,  $\alpha \approx 0.83$  for Li.

## FINITE PULSE LENGTH AND DIFFRACTION

So far the total energy of the pulse (or its total duration for the fixed intensity) has had no influence on the process. Obviously, this holds up to a distance  $L'$ , where the total absorption or conversion of the pump pulse occurs. When there is no photoionization of ground-level atoms by the Stokes pulse, as in Li, every atom on the pulse path absorbs one photon from the pump either because of photoionization or because of the Raman process. The total pump energy flux therefore is

$$J_p(z) = J_p - N\hbar\omega_p z. \quad (24)$$

In He pump absorption may, in principle, decrease because some atoms are ionized by Stokes photons. Numerical simulation, however, shows that this decrease is negligibly small. Therefore, the approximate analytical models described above hold up to and including a distance  $z$  satisfying

$$z \leq L' \approx J_p/N\hbar\omega_p, \quad (25)$$

where  $L'$  is the total absorption length of the pump pulse. For larger  $z$ ,  $J_s(z) \approx J_s(L')$  in Li and  $J_s(z) \approx J_s(L')\exp(-z/l_s)$  in He.

As is stated above, our analysis is based on a plane-wave approximation. Although we did not investigate the influence of diffraction losses in detail, it is clear

that, at least for small gain  $G$ ,<sup>3</sup> the useful cell length  $L$  will not exceed  $\sim b$ , where  $b$  is the confocal parameter of the Stokes beam. Therefore, assuming that  $b = L$  and stipulating that the optimal situation corresponds to  $L = L'$ , we obtain, using relation (25) (note that now  $J_p = 2W_p/\lambda_s L$ , where  $W_p$  is the pump-pulse energy), the optimal cell length  $L_{\text{opt}}$ , for both Li and He:

$$L_{\text{opt}} = (2\lambda_p W_p / \lambda_s N h c)^{1/2}. \quad (26)$$

In Li the maximum Stokes output energy is  $W_s(L_{\text{opt}}) \approx W_s^{\text{eff}} \exp(GL_{\text{opt}})$  for  $L_{\text{opt}} \leq z_1$  and  $W_s(L_{\text{opt}}) \approx \alpha N \hbar \omega_s (L_{\text{opt}} - z_2)$  for  $L_{\text{opt}} > z_2$ . For a state-of-the-art x-ray laser pulse energy of  $\sim 3 \times 10^{-4}$  J the optimal focusing for  $N = 10^{18} \text{ cm}^{-3}$  is  $b = L \approx 5.65$  cm and the total exponential gain at the end of the cell  $GL \approx 23$ . The effective initial Stokes energy needed to reach saturation at the end of the cell is  $\sim 10^{-14}$  J. To achieve saturation without the seeded Stokes pulse, i.e., using only spontaneous emission at the Stokes frequency, we must increase the energy of the pump by approximately 1 order of magnitude.

In He the maximum energy conversion  $\eta_{\text{max}}$  is obtained when  $L_{\text{opt}} = z_{\text{cr}}$  and is given by<sup>26</sup>

$$\eta_{\text{max}} = W_s(L_{\text{opt}})/W_p \approx \lambda_s z_{\text{cr}} J_{\text{max}}/2W_p. \quad (27)$$

Thus for a given initial pump energy there is an optimal atomic-number density that is required for maximum efficiency, and, vice versa, for a given atomic-number density there is an optimal pump energy. Either can be calculated from the condition  $L_{\text{opt}} = z_{\text{cr}}$  with relation (19) and Eq. (26). Assuming, for example, that  $W_s(0)/W_p = 10^{-12}$ , we obtain that for  $N = 10^{19} \text{ cm}^{-3}$  the optimal pump energy is  $\sim 2.2 \times 10^{-3}$  J with a resulting Stokes output of  $\sim 1.6 \times 10^{-5}$  J from an  $\sim 1.5$ -cm cell.

## CONCLUSIONS

In conclusion, we have proposed configurations for observing x-ray SERS in initially neutral gases (He and Li). Numerical solution of the Maxwell-Bloch equations with photoionization taken into account shows that considerable conversion efficiencies can be achieved by operation in a high-pump-energy regime even when the total photoionization is much faster than the pulse duration. We also developed an approximate analytical model and found optimal parameters for focused beams. We showed that short high-energy Stokes pulses, as well as sharpened Se x-ray laser pulses, can be obtained in the process. In Li the x-ray SERS generates a strong precursor field. We found that coherent x-ray SERS can significantly inhibit the photoionization induced by x-ray radiation in both gases.

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