X-ray stimulated Raman scattering in Li and He

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We propose to use initially neutral Li and He gases to observe x-ray stimulating Raman scattering. We solve the Maxwell–Bloch equations for the Raman scattering in the presence of a strong photoionization atomic depletion and show that very high conversion efficiencies can be obtained, although the required intensities are significantly high to cause very fast photoionization.

Among other possible nonlinear optical effects in the x-ray domain, x-ray stimulated electronic Raman scattering (SERS) has a special significance since it provides an opportunity to attain coherent radiation with new frequencies by using (initially) neutral atoms instead of preionized plasmas.[1] In this letter we present our first results on the SERS feasibility in nonionized gases.[2] Any realistic proposal for observing x-ray SERS has to address two major questions: (1) How to provide necessary resonant enhancement? (2) Is it possible to obtain an observable Stokes output in a regime where atoms are significantly, or even totally, ionized by the pump pulse in a very short time?

Concerning the first question we note that, since x-ray laser photon energy (~50–300 eV) is much larger than the binding energy of optical electrons in any neutral atom, no resonances to optical transitions are possible. Instead, we suggest to make use of the so called core-excited, or multiple-excited, atomic states with at least one of the inner electrons excited.[3] In particular, some double-excited levels of He and Li atoms are resonant to the powerful Se^{+24} 20.9 nm x-ray laser.[4,5] Two transition schemes were consider-ed: (i) He: $1^1S \rightarrow 2s2p\,^1P \rightarrow 2^1S$ (the expected Stokes wavelength 32.2 nm), and (ii) Li: $1s^22s \rightarrow [1s(2s2p)^1P]^2P \rightarrow 1s^23s^2S$. (the expected Stokes wavelength 22.3 nm). To estimate the pump intensities required to obtain a significant Stokes output and the extent of the photoionization influences on the process we first calculated the small signal cw Raman gain and the photoionization cross sections. The small signal cw exponential gain in the situation of resonant enhancement, both in He and in Li, can be calculated[6] as $G_{cw}=I_pNg=I_pN\hbar\omega_sK^2/2\Gamma$ with

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

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, Γ is the width of the Raman transition, ω_s (ω_p) is the Stokes (pump) frequency, Ω_{31} (Ω_{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 intermediate state. For $\Gamma=\Gamma_D$ (Doppler broadening) and N in units of 10^{18} cm^{-3}, we obtain[7–13] for He: $g=2\times10^{-12}$ W^{-1} cm, and for Li: $g=1.5\times10^{11}$ W^{-1} cm. The photoionization process in He is dominated by the direct photoionization of the ground state atoms with the cross sections for Stokes and pump being $\sigma_s\approx\sigma_p\approx10^{-18}$ cm^2. In Li the direct photoionization is negligible, as compared to the absorption to the intermediate autoionizing double-excited level. The cross sections of the latter can be calculated as[5]

$$\sigma_p=2\pi r_ecf_{13}\Gamma_3(\Omega_{31}-\omega_p)^{-2},$$
$$\sigma_s=2\pi r_ecf_{23}\Gamma_3(\Omega_{32}-\omega_s)^{-2}, \tag{2}$$

for the pump and the Stokes radiation, respectively. Here $\Gamma_3=2.6$ meV is the autoionization width of the intermediate level. Using appropriate data we find that in Li, $\sigma_p=7\times10^{-19}$ cm^2 and $\sigma_s=4\times10^{-21}$ cm^2. Thus the characteristic absorption length is $(N\sigma_p)^{-1}\approx1$ cm for $N=10^{18}$ 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} W/cm^2. Such intense radiation would, however, fully ionize the gas much faster (e.g., ~1 ps for pump intensity of 10^{13} 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.

To the best of our knowledge, a coherent SERS in the presence of the ionization-related atomic depletion has never been considered. In general, SERS is governed by coupled Maxwell–Bloch equations,[14,15] which in our case have to be modified to include the effect of photoionization. We assume plane-wave pulses traveling with the same phase and group velocity c (which is justified for the typical gas pressure in the x-ray experiments) and represent the pump (E_p) and the Stokes (E_s) fields as $E_{p,s}=\{E_{p,s}\exp[i(\omega_{p,s}t-k_{p,s}z)]+\text{c.c.}\}/2$, where $E_{p,s}$ are slow-varying complex field envelopes and $k_{p,s}=\omega_{p,s}/c$. Since the excited Raman levels in He and Li have lifetimes of a few microseconds[16] and nanoseconds,[17] respectively, we neglect both the inhomogeneous and homogeneous broadening. We also neglect linear nonresonant susceptibilities, $\chi_1(\omega_{p,s})\approx\chi_2(\omega_{p,s})\approx0$, which is also justified in the x-ray domain.

The Maxwell–Bloch equations can then be written in the retarded frame, $\tau=t-z/c$, $\xi=z$, in terms of ground and exited level populations, ρ_{11} and ρ_{22}, field intensities $I_{p,s}=|E_{p,s}|^2$ and slow-varying envelope, ρ, of the nondiagonal elements of density matrix defined as $\rho_{21}=\rho_{12}^*=i\rho\exp[i(k_p-k_s)z-i\omega_0t]$, as[2]

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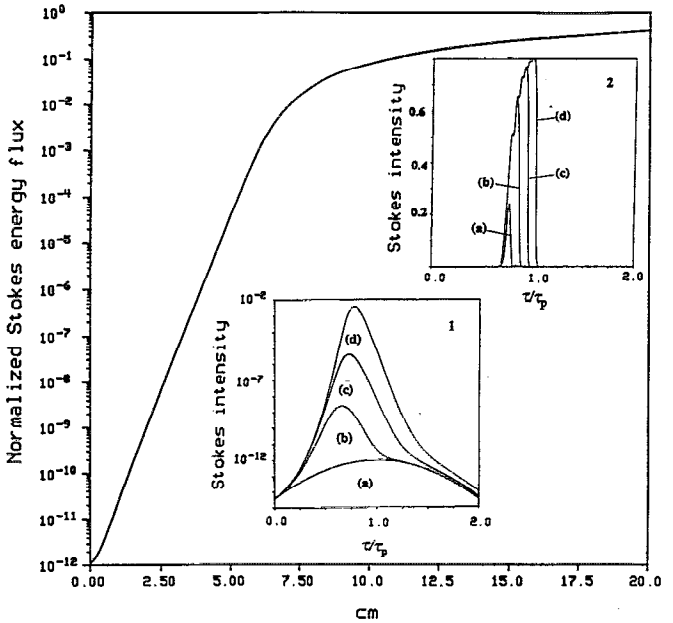

FIG. 1. Normalized Stokes energy flux $J_s(z)/J_p$ in Li as a function of the cell length z. Insets: Normalized Stokes intensity $I_s(z,\tau)/I_{p\ \max}$ as a function of normalized retarded time τ/τ_p in (1) exponential region; curves: (a) $z=0$ cm; (b) $z=2$ cm; (c) $z=4$ cm; (d) $z=6$ cm; in (2) linear region; curves: (a) $z=8$ cm; (b) $z=12$ cm; (c) $z=16$ cm; (d) $z=20$ cm.

$$\partial\rho_{jj}/\partial\tau=(-1)^j U-w_j\rho_{jj}\quad (j=1,2), \tag{3a}$$

$$\partial\rho/\partial\tau=U(\rho_{11}-\rho_{22})/2-(w_1+w_2)\rho/2, \tag{3b}$$

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

$$(i=p,s) \tag{3c}$$

where $U^2=K^2 I_p I_s$, $S_p=-S_s=-1$, $w_j=\beta_j^p|E_p|^2+\beta_j^s|E_s|^2$ and 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 $\sigma_{p,s}\approx 10^{-18}$ cm^2, while in Li, $\beta_1^s=\beta_2^p\approx 0$, $\beta_2^s\approx\sigma_s\epsilon_0 c/2\hbar\omega_s$, and $\beta_1^p\approx\sigma_p\epsilon_0 c/2\hbar\omega_p$ [for $\sigma_{p,s}$ see Eq. (2)].

Figure 1 shows typical results of a numerical solution of Eq. (3) for Li. Normalized Stokes pulse energy flux $J_s(z)/J_p$ is shown as a function of the cell length z for $N=10^{18}$ cm^{-3} (corresponding to the pressure of $\sim$0.1 atm at $T=800$ K), the initial pump pulse energy flux $J_p=2.3\times10^2$ J/cm^2, and initial normalized energy $J_s(0)/J_p=10^{-12}$. In our calculations we assume that the seeded Stokes pulse has the same (i.e., Gaussian) shape and duration as the pump pulse. Figure 1 clearly shows two distinct stages of this Raman process, namely the exponential growth and the saturation. The numerical results for He are qualitatively the same except for the limiting behavior in the saturation region. While in Li, as seen in Fig. 1, the Stokes energy growth gradually becomes linear, it saturates completely in He resulting in a stable, soliton-like output Stokes pulse, which will be addressed by us in detail elsewhere. Insets in Fig. 1 show normalized Stokes intensity $I_s(z,\tau)/I_{p\ \max}$ as a function of normalized retarded time τ/τ_p, where τ_p is the pump pulse width, at different z in both the exponential and linear region. In the exponential region the Stokes pulse width is constant and its peak coincides with the leading edge of the pump pulse, whose velocity, v (not to be confused with the pump group velocity) is limited by the photoionization, and thus is lower than c; in the exponential region it can be approximately estimated as

$$v\approx(1/c+N/N_{\rm ph})^{-1}, \tag{4}$$

where $N_{\rm ph}=|E_p|^2_{\max}\epsilon_0 c/2\hbar\omega_p$ is the pump photon flux. In the saturation region, the Stokes pulse intensity is almost constant while the pulse width increases linearly with z. Note that the conversion efficiency [i.e., $J_s(L)/J_p$] for Li reaches almost unity for sufficiently long cell.

Based on our approximate analytical model of the process,[2] the Stokes energy flux in the exponential region can be described as

$$J_s(z)\sim\exp[(G-1/l_s)z], \tag{5}$$

where $G=K^2(N^2\hbar^2\omega_s\omega_p/\epsilon_0^2 c^2)l_p$, $l_p=\gamma_p(N\sigma_p)^{-1}$, and $l_s=\gamma_s(N\sigma_s)^{-1}$. The numerical factors $\gamma_{p,s}$ were determined from the comparison with the numerical results. The best fit in both gases was obtained with $\gamma_p=0.77$. In Li, γ_s is irrelevant since one can neglect $(l_s)^{-1}$ [for example, $N=10^{18}$ cm^{-3} gives $(N\sigma_s)^{-1}=300$ cm]. In He, the best fit was obtained with $\gamma_s=2.5$. Eq. (5) shows that when the full photoionization occurs faster than the pump pulse duration the amplification of the Stokes signal is possible only if $l_s G>1$. In general case, defining a Raman cross section as $\sigma_R\equiv(\gamma_p\gamma_s G/N^2 l_p)^{1/2}$, and noting that, due to the definition of G, it depends only on spectroscopic parameters of the atom, this condition can be expressed in a purely spectroscopic form

$$\sigma_R^2=\gamma_p\gamma_s K^2\hbar^2\omega_s\omega_p/\epsilon_0^2 c^2>\sigma_p\sigma_s. \tag{6}$$

For not very large pump energy, i.e., when $\tau_p<(w_j)^{-1}$, one can neglect the "photoionization terms w_j in Eq. 3a and 3b, which results in almost standard equations for coherent Raman scattering.[14,15] In the small area approximation,[14] we expect then 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 Raman process increases too, the photoionization intensifies and gets 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 exponential gain of $\sim$3.6 at $z=l_p$ and, under the same conditions as in Fig. 1, the conversion efficiency is $\sim6\times10^{-9}$. This is in striking contrast to the case depicted in Fig. 1 corresponding to $\tau_p\gg\omega_1^{-1}$ in which case the conversion efficiency approaches unity. It is therefore clear, that increase in the pump intensity to the point $\tau_p\gg(w_1)^{-1}$, can result in an enormous increase not only in the absolute Stokes energy output, but also in the conversion efficiency.

Both in Li and in He, the saturation sets in because of Raman-related atomic depletion[6] causing the Raman ampli-

fication factor G in Eq. (5) to decrease with increasing Stokes intensity. In Li, this process stops when the the Stokes intensity becomes comparable to that of the pump (see Fig. 1). After that the Stokes energy growth becomes linear $J_s(z) \sim \alpha N \hbar \omega_s$, where $\alpha = \rho_{22}(z,\infty) = 0.83$ is the excited level population after the passage of the pulse. In He, the Stokes energy growth saturates completely, $J_s(z) = J_{\max} \approx 3$ J/cm^2, when the balance is reached between the amplification and absorption [i.e., $G \approx (l_s)^{-1}$]. Our numerical and approximate analytical results show that both α and $J_{\max}$ are spectroscopic constants independent of atomic number density, pump intensity, and the cell length.

Apart from the saturation described above, it is obvious that any growth in the Stokes energy can be sustained only up to a distance L_{ion} at which the pump energy is totally depleted due to photoionization, i.e.,

$$z \leq L_{\text{ion}} \approx J_p / N \hbar \omega_p . \tag{7}$$

Although we used plane-wave approximation, it is clear that, at least for small gain G, the useful cell length L will not exceed the confocal parameter[6] of the Stokes beam, b. Our analysis shows that the Stokes energy output reaches its maximum when $L = L_{\text{ion}}$. Thus, assuming $L = b$, the optimal cell length is

$$L_{\text{opt}} = (2 \lambda_p W_p / \lambda_s N h c)^{1/2}, \tag{8}$$

where W_p is the pump pulse energy. In Li, for the state-of-the-art x-ray laser pulse energy of $\sim 3 \times 10^{-4}$ J the optimal focusing for $N = 10^{18}$ cm^{-3} is $b = L \approx 5.65$ cm and the total exponential gain at the cell end is $GL \approx 23$.

In conclusion, we showed that the x-ray pump required for significant Stokes generation, causes the full photoionization to occur much faster than the pump pulse duration. Despite this fact, x-ray SERS in (initially) neutral Li and He may result in high conversion efficiencies.

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