NEW METHOD OF INCREASING THE EMISSION FREQUENCY OF HIGH-POWER LASER PULSES

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Usually the active media used for stimulated Raman scattering (SRS) are molecular gases or liquids in thermodynamic equilibrium. Most molecules are then in the ground state, and a series of Stokes components $\omega_{-1} = \omega_0 - \Omega$, $\omega_{-2} = \omega_0 - 2\Omega$) etc. is generated in the SRS, i.e., the frequency ω_0 of the incident radiation is decreased by one, two, etc. vibrational quanta of the molecule (Ω). Such a lowering of the frequency can be realized with a near-unity photon conversion efficiency [1].

It is quite obvious that in the case of SRS in an inverted medium, the opposite process, an increase of the frequency, will take place. For the first anti-Stokes component we have $\omega_1 = \omega_0 + \Omega$, for the second $\omega_2 = \omega_0 + 2\Omega$, etc. This raises naturally the question of finding an inverted system in which an appreciable increase of the radiation frequency can be effected, with a high conversion efficiency. In other words, we seek an inverted medium in which a sufficiently high gain can be obtained at reasonable values of the pump intensity. Among the active media used in gas lasers, one of the most suitable from this point of view is excited iodine. Photodissociation of the compounds CF_3I and C_3F_7I produces an inversion density $N = (3 - 6) \times 10^{17}$ cm^{-3} for the atomic iodine transition $5\ ^2P_{1/2} - 5\ ^2P_{3/2}$ ($\lambda = 1.315\ \mu$, $\hbar\Omega = 0.95$ eV) [2].

The gain for SRS with polarized pumping is given by

$$k = 4\lambda^2 \frac{\frac{d\sigma}{d\theta} I_0 N}{\hbar\omega_0 (\Delta\omega + \Delta\omega_p)}, \qquad (1)$$

where λ is the wavelength of the scattered radiation, I_0 is the intensity of the pump radiation, $\hbar$ is Planck's constant, $d\sigma/d\theta$ is the differential cross section (with respect to the solid angle) of the Raman scattering (RS), $\Delta\omega$ is the width of the RS line, and $\Delta\omega_p$ is the width of the pump spectrum. The cross section of RS by atomic iodine can be estimated with sufficient reliability from the general formulas of [3, 4]. Since both iodine states of interest to us are far enough from other levels and from the continuous spectrum, the summation of the matrix elements over the virtual states can be carried approximately with the aid of the sum rule. At the neodymium-laser wavelength ($\lambda_0 = 1.06\ \mu$), such an estimate yields $d\sigma/d\theta \simeq 1 \times 10^{-28}$ cm^2/sr, which is approximately 3×10^3 times larger than the cross section of RS per molecule in liquid nitrogen. This means that in the case of anti-Stokes SRS by inverted iodine we can count on obtaining the same gain as in ordinary Stokes SRS. The wavelengths of the anti-Stokes components are $\lambda_1 = 0.59\ \mu$, $\lambda_2 = 0.41\ \mu$, $\lambda_3 = 0.31\ \mu$, $\lambda_4 = 0.25\ \mu$, and $\lambda_5 = 0.21\ \mu$. With decreasing wavelength, the RS cross section increases in proportion to λ^{-3}, so that conversions into higher anti-Stokes components are realizable, i.e., the emission frequency can be greatly increased.

Let us examine in greater detail the conversion into the first anti-Stokes component. Unlike the ordinary Stokes scattering, in our case the saturation of the inversion of the iodine transition is important. The number of anti-Stokes photons can in any case not be larger than two-thirds of the inversion stored in the volume (with allowance for the statistical weights of the iodine levels). At typical values of the inversion density $N \approx 3 \times 10^{17}$ cm^{-3} we can store in one tube of ~30 cm length[1)] a useful inversion of ~10^{19} per square centimeter of cross section, i.e., we can convert into the first anti-Stokes components 10^{19} photons/cm^2 (~1 J/cm^2).

The iodine line width is determined by the Doppler effect and by collisions with the molecules of the active medium. Depending on the pressure, in ranges from the Doppler value (0.1 cm^{-1}) to 0.5 cm^{-1} [5]. If the pump line width does not exceed 0.01 cm^{-1}, we can obtain a gain $K > 0.01$ cm^{-1} for $I_0 \approx 2 \times 10^7$ W/cm^2, which is quite sufficient to produce lasing in the resonator, and furthermore simultaneous lasing at two or even three anti-Stokes components (λ_1, λ_2, λ_3). To obtain high efficiency and high radiation directivity, a generator + amplifier system is most suitable. The gain required in the amplifier is $k \approx 0.3 - 0.5$ cm^{-1}. Calculation shows that the efficiency, in terms of the number of photons, exceeds 70% in such a system with the following parameters: $I_0 > 6 \times 10^9$ W/cm^2, $\Delta\nu = \Delta\omega/2\pi c = \Delta\omega_p/2\pi c = 0.1$ cm^{-1}, and with a pump pulse duration $\tau < 0.3$ nsec. By using the radiation of the first Stokes component as the pump of the next conversion stage, we can effect transformation into the second anti-Stokes component, etc.

Thus, SRS by excited atomic iodine enables us to convert high-power radiation from a neodymium laser into the ultraviolet region of the spectrum with quite high efficiency (the photon efficiency for $\lambda_3 = 0.31$ μ exceeds 30%[2)]; the pulse energy at λ_3 exceeds the energy of the initial pump pulse).

The proposed method of increasing the radiation frequency, in contrast to crystalline harmonic generators, does not call for high directivity of the pump radiation. In addition, the use of a gaseous active medium allows us to work with radiation intensities greatly exceeding the crystal damage threshold. The use of the proposed method of increasing the frequency therefore seems quite promising for laser heating of a plasma.

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[1)] The tube length is limited by self-excitation of generation at the proper transition of iodine with $\lambda = 1.315$ μ.

[2)] In the case of excess inversion, the photon efficiency can approach unity.