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A NEW METHOD FOR UPCONVERSION OF MONOCHROMATIC RADIATION

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SUMMARY

Frequency upconversion by means of stimulated Raman scattering is examined. A monochromatic beam is scattered into the anti-Stokes wave in a medium with an inverted population of the pair of energy levels involved. A method is proposed in which the beam from a laser tuned to the desired anti-Stokes frequency provides an initial stimulating wave of sufficient intensity to ensure the desired Raman scattering dominates competitive processes.

INTRODUCTION

Consider a medium with various excitations, either localized at the molecules, atoms, and so forth or distributed in collective motions such as phonons. We focus our attention on (1) a certain type of excitation from which a radiative transition can occur with emission of a photon of energy $E = h\nu$, (2) a particular spontaneous transition of this kind whose emitted photon has propagation vector (in the medium) $\vec{k}$, $k = (2\pi n/c)\nu$, and (3) a wave of photons $\vec{k}$ associated with the stimulated counterpart of this spontaneous emission. At any location $\vec{r}$, the transition probability per unit time for stimulated emission into the mode $\vec{k}$ is proportional to $N_{\vec{k}}(\vec{r})$, the number of photons per unit volume in the mode before emission occurs. If the wave $\vec{k}$ experiences uniform radiation and excitation conditions equivalent to a constant positive gain $g(\nu)$ and the gain exceeds a certain threshold value, then $N_{\vec{k}}(\vec{r})$ will grow exponentially with distance along $\vec{k}$ as the wave traverses the medium. Such is the case in a region where a stimulated wave starts to build up from the noise level of the spontaneous process. If the threshold for growth of the stimulated wave $\vec{k}$ is attained before the threshold for competing processes, then once the wave develops it will increase rapidly and dominate the other processes.

Alternatively, when the number of quanta in the mode $\vec{k}$ is large compared to other modes, the transition probability for the particular stimulated transition greatly exceeds that for other stimulated processes. If there is present initially a sufficiently intense wave of stimulating radiation in the mode $\vec{k}$, the initial rate of growth of this wave will be large enough to ensure that stimulated emission into the mode $\vec{k}$ via the given process is dominant. A laser beam is ideal for supplying the required initial intensity of stimulating radiation. The beam from a relatively small laser will be adequate in many cases and the coherence of the laser beam will mediate the coherence of the ensuing stimulated emission.

APPLICATIONS AND RESULTS

We now apply these principles to obtain frequency upconversion of an intense monochromatic beam of radiation ν_L by stimulated Raman scattering (SRS) into an anti-Stokes frequency. A schematic configuration is shown in figure 1. In the medium of the converter cell, the active molecules have an inverted population of the energy levels

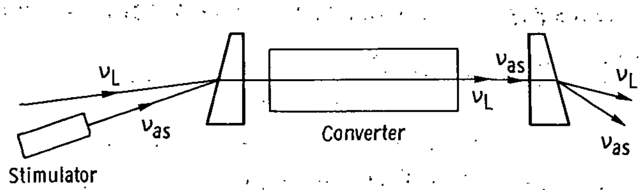

Figure 1. - Schematic configuration for frequency upconversion using stimulated Raman effect. In the arrangement shown, intense beam ν_L and stimulating beam ν_{as} enter converter cell in parallel.

$E_b > E_a$, that is, $N_b > N_a$, where N_b is the number of molecules per unit volume in the upper energy state $|b\rangle$, and so forth. In the radiative process of interest, a wave having frequency ν_L and propagation vector $\vec{k}_L$ is incident on a molecule in the upper energy state $|b\rangle$, simultaneously with a wave at the anti-Stokes frequency $\nu_{as} = \nu_L + \nu_{ba}$, having the propagation vector $\vec{k}_{as}$, where $h\nu_{ba} = E_b - E_a$. The particular transition considered is the Raman scattering in which a photon $\vec{k}_L$ is absorbed from the first wave and a photon $\vec{k}_{as}$ is emitted into the second wave, while the molecule contributes the required energy $h\nu_{ba}$ in going to the lower state $|a\rangle$ (ref. 1). The population inversion of the two molecular levels is presumed to be large enough to make the converter cell a high gain amplifier for this process.

The initial stimulating wave is supplied by the stimulator, a tunable laser controlled to oscillate at the desired anti-Stokes frequency ν_{as}. The intense beam ν_L may also originate from a laser, but is only required to be monochromatic and need not necessarily be coherent. For efficient upconversion of the beam ν_L, its frequency width should not exceed the frequency width of the stimulating wave ν_{as}. In principle, a radiation beam from an incoherent source, if sufficiently intense in such a frequency width, could serve as the wave ν_L and in the process be upconverted into a coherent beam.

Any Raman scattering into a Stokes component (e.g., by exciting molecules from the ground state to a higher energy state) has positive gain when the population of the two states involved is normal. Hence the converter cell is potentially an ordinary Raman laser with the beam ν_L as the pump or a parametric amplifier coupling the beam to some other excitation. Consider the Stokes wave with the largest gain coefficient, that is, with the strongest Raman scattering from the beam ν_L. Let $h\nu_M$ be the energy increase of the particular excitation involved. In the absence of the laser beam ν_{as}, a Stokes wave $\nu_L - \nu_M$ will grow by SRS and dominate whenever the beam ν_L exceeds a certain threshold intensity. The incident coherent beam ν_{as} serves two purposes. It stimulates the Raman scattering of the beam ν_L into the desired anti-Stokes wave ν_{as}, and thereby prevents the growth of stimulated processes with competitive thresholds. It also imposes its coherence on the induced anti-Stokes emission.

Let us look at the operation of the converter under the conditions of single-pass conversion of the beam ν_L and uniform inversion along the converter. Consider the case where the beam ν_L is pulsed and the pulse length is small compared to the converter length. Then each pulse experiences over its extent an effective population inversion $N_b - N_a$ that is independent of position z along the converter cell. The stimulating laser beam ν_{as} may be pulsed as well, so long as the pulses of the two beams spatially overlap. When the anti-Stokes beam ν_{as} is directed along the length of the converter, the average (over neighboring modes) number $N(z, \nu)$ per unit volume of anti-Stokes photons at frequence ν varies with z according to (ref. 1)

$$\frac{dN(z, \nu)}{dz} = (N_b - N_a)\left(\frac{c^3}{\nu^2 n^3}\,\frac{d\sigma_{sp}^R}{d\Omega}\right) h(\nu)\; N_L(z)\; N(z, \nu) \tag{1}$$

The Lorentzian $h(\nu) \equiv 2\Gamma_{ab}\Big/\left[4\pi^2(\nu_L + \nu_{ba} - \nu)^2 + \Gamma_{ab}^2\right]$, with the half-width at half-maximum Γ_{ab} appropriate to the pair of levels, represents the line-shape for the two induced processes, stimulated emission and absorption of the anti-Stokes photons. In the beam ν_L, whose frequency width we ignore, the number of photons per unit volume at z is $N_L(z)$. The term proportional to N_b describes the SRS from the beam ν_L into the anti-Stokes wave. The term proportional to N_a corresponds to an absorption of anti-Stokes photons at ν (with excitation of molecules from the lower to the upper energy state) due to SRS into the beam ν_L, which is now the stimulating wave. The quantity $d\sigma_{sp}^R/d\Omega$ is the differential cross section for the spontaneous Raman scattering of photons ν_L into the anti-Stokes frequency ν_{as}, and $n(\nu)$ is the index of refraction in the medium. The function $N(0, \nu)$ centered at $\nu = \nu_{as}$ is the frequency distribution of the incident laser beam ν_{as}.

The higher gain at the line center will preserve or even narrow the spectral distribution of the anti-Stokes wave as it traverses the converter. Taking the frequency width of $N(z, \nu)$ to be small compared to Γ_{ab} permits us immediately to sum equation (1) over ν and obtain

$$\frac{dN(z)}{dz} = (N_b - N_a)\left(\frac{c^3}{\nu_{as}^2 n_{as}^3} \frac{d\sigma_{sp}^R}{d\Omega} \frac{2}{\Gamma_{ab}}\right) N_L(z)\, N(z) \tag{2}$$

where $N(z) = \int N(z, \nu)\, d\nu$.

The geometry in figure 1, where the incident beams are parallel, is most efficient for overlap of the pulses. If the attenuation of the beam ν_L in the converter is due entirely to photon exchanges with the anti-Stokes wave, we have $N_L(z) = N_L(0) - [N(z) - N(0)]$. This enables us to write equation (2) in the form $dy/dz = g(y - y^2)$, where $y(z) = N(z)/[N_L(0) + N(0)]$ and

$$g = (N_b - N_a)\left(\frac{d\sigma_{sp}^R}{d\Omega}\right)\left(\frac{c^3}{\nu_{as}^2 n_{as}^3}\right)\left(\frac{2}{\Gamma_{ab}}\right)\left[N_L(0) + N(0)\right] \tag{3}$$

is an effective gain coefficient. At the value z_1 where $y(z_1) = 1/2$, the anti-Stokes wave attains its maximum rate of growth. The integral of the differential equation can be written

$$y(z) = y_o e^{gz} \left(1 - y_o + y_o e^{gz}\right)^{-1} \quad \text{and} \quad y_o \equiv y(0) = \frac{N(0)}{N_L(0) + N(0)}$$

When $y_o << 1$, the fractional conversion is $y(z') = p$ at $z' = g^{-1}\left\{\ln[p/(1 - p)] - \ln y_o\right\}$.

For an incident ratio $y_o = 10^{-5}$, a gain $g \sim 0.1$ centimeter^{-1} gives 90 percent conversion in a length of 135 centimeters. Such a magnitude of the gain coefficient is obtained in Raman lasers pumped at peak intensities ($\lambda_L = 6943$ Å) of 5 to 50 megawatts per square centimeter (condensed media to high density gases)(ref. 2). The expression for the gain coefficient of a Raman laser has the same form as equation (3) with the inversion term written as $N_a - N_b$ where N_a and N_b are the normal occupation numbers of the ground state and excited state, respectively. For a Raman laser, when

$kT << h\nu_{ab}$, N_b is small and $N_a - N_b \approx N_o$, the number of active molecules per unit volume. From the pump intensities and inversion for Raman lasers at $g = 0.1$ centimeter^{-1}, we infer that a converter cell with the same gain and a population inversion of $10^{-3}\ N_o$ in a condensed medium, requires an intensity of about 5×10^9 watts per square centimeter when the Raman cross section, and so forth are comparable. Thus the up-conversion of short duration pulses requires intensities that are high but readily attainable.

Besides the incident intensity, other factors in the gain should be optimized to counteract the small cross section for Raman scattering (a second-order process). The cross section is generally isotropic and largest for a totally symmetric vibration. With a polarized incident wave, the spontaneous Raman scattering is maximum for parallel polarization of the scattered wave. Thus parallel polarization of the incident beams is favored. Enhanced population inversion will result from "four-level" operation, that is, from a lower state with rapid decay and small normal population. The threshold for amplified spontaneous emission between the pair of levels imposes an upper limit to inversion. But this threshold inversion greatly exceeds that for ordinary laser oscillation in the medium, due to the absence of feedback and the generally small direct radiative probability between a pair of Raman active levels. The parallel beam geometry in figure 1 has a further advantage if self-focusing occurs, since anomalously high gain in the beam direction usually accompanies this effect.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, July 11, 1973,
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