Characteristics of a Raman Laser Excited by an Ordinary Ruby Laser

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Abstract—The stimulated Raman effect of benzene has been observed using an ordinary (nongiant) ruby laser. The build-up of oscillation at the ν_2, $2\nu_2$, $3\nu_2$ and $4\nu_2$ Stokes lines and also at the first ν_1 Stokes line have been observed. The threshold exciting power for laser action in the ν_2 Stokes line has been measured to be 9.5 kW. A rate equation for the Raman laser has been given, and the total scattering cross section for the ν_2 Raman line of benzene has been determined as $\sigma = 0.46 \cdot 10^{-28}$ cm². The estimation based on the results of this investigation indicates that it is possible to construct a Raman laser of benzene using an Ar gas laser.

INTRODUCTION

IT WAS FOUND by Woodbury, *et al.* [1], [2], [3] that Raman active material in an optical cavity builds up optical radiation at one or more Raman shifted frequencies when it is excited with strong radiation generated by a Q-switching (giant pulse) laser.

Recently, they also found that stimulated Raman scattering can be induced at the ν_2 (992 cm⁻¹) Stokes line of benzene even in an ordinary (nongiant) laser [5]. We have also found independently that the stimulated Raman radiation can be built up not only at the ν_2 Stokes line, but also at the $2\nu_2$, $3\nu_2$, $4\nu_2$, and the ν_1 (3064 cm⁻¹) Stokes lines of benzene using ordinary ruby lasers.

This is in one sense an improvement of the efficiency of the stimulated Raman scattering of as much as several hundred times, and it is interesting to investigate the detailed characteristics of this kind of Raman laser.

The results of our experimental investigation and analysis of the Raman laser excited by an ordinary ruby laser will be described in the following parts of this paper.

EXPERIMENTAL PROCEDURES AND RESULTS

A ruby rod which is 6 mm in diameter and 75 mm in length has been used in the following experiments, though we have been able to observe the stimulated Raman effect using other ruby rods.

Both ends of the ruby rod are polished to $\frac{1}{10}\lambda$, parallel to two seconds of arc, and a dielectric antireflection coating is deposited on one end and a multilayer dielectric coating of 99.6 per cent reflectivity is deposited on the other end. The concentration of Cr³⁺ ion is 0.04 per cent, and the rod axis is perpendicular to the optical axis of the ruby crystal.

A plane mirror of 95 per cent reflectivity and the end

of the ruby rod with high reflectivity make an optical resonator for both the ruby and the Raman laser. A Raman cell which has antireflection coated optical flats as windows and a 95-mm effective length is inserted in the optical resonator.

Benzene has been used in the following experiments as the Raman active liquid.

The spectrum of the output beam has been examined with a grating spectrometer. A typical spectrum of the output beam for several values of the input energy is shown in Fig. 1. The spectrum of the output radiation was observed using the same grating spectrometer after removing the slit and the collimator lens as shown in Fig. 2. Since the output beams of the lasers are highly parallel, such a method has been found to be useful; we can measure the angular divergence of the output beam of each Stokes component. The angular divergence of the ν_2, $2\nu_2$, and $3\nu_2$ Stokes lines determined with this method are $2.5 \cdot 10^{-3}$ rad, $2.5 \cdot 10^{-3}$ rad, and $4 \cdot 10^{-3}$ rad, respectively.

The threshold input electric energy to build up stimulated Raman radiations at the ν_2, $2\nu_2$, $3\nu_2$, and ν_1 Stokes lines of benzene has been found to be $1.3E_{th}$, $2.3E_{th}$, $2.5E_{th}$, and $2.5E_{th}$, respectively, where E_{th} is the threshold input energy to build up the ruby laser oscillation. The $4\nu_2$ Stokes line can also be built up when the alignment of the optical system is made very carefully and the input energy is higher than $2.7E_{th}$.

Near field patterns of the Raman laser output have been photographed using a piece of infrared pass filter which transmits Stokes lines but does not pass the ruby laser output. Two near field patterns of the Raman laser at exciting input energies slightly higher and much higher than the threshold value are shown in Fig. 3. It is shown in Fig. 3 that the Raman laser oscillation is composed of several "filaments," each of which is an independent optical resonator. The average cross section of the filaments when the input electric energy is close to the threshold value has been determined to be about $2 \cdot 10^{-3}$ cm².

The threshold exciting power of the incident ruby laser radiation to build up the stimulated Raman radiation has been measured from the dual beam oscilloscope displays of the Raman and the ruby laser spikes. Typical waveforms of the ruby and the Raman laser spikes are shown in Fig. 4. A high-current biplanar phototube with an S 1 cathode and an infrared pass filter have been used to isolate and detect the Stokes components. The same type of phototube with an S 20
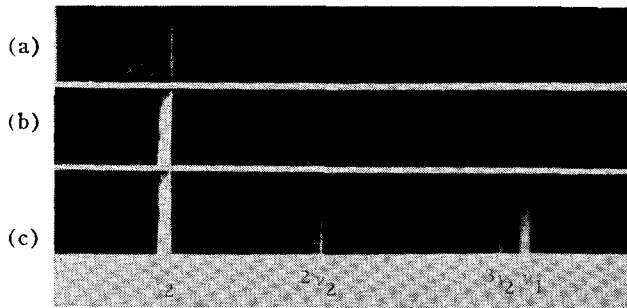
Fig. 1. Output spectrum of the Raman laser at various input energies, (a) 1.3 E_{th}. (b) 2.3 E_{th}. (c) 2.5 E_{th}.

Fig. 2. Spectrum of the Raman laser output. The slit and the collimator are removed from the grating spectrometer, and the camera is focused at infinite distance.
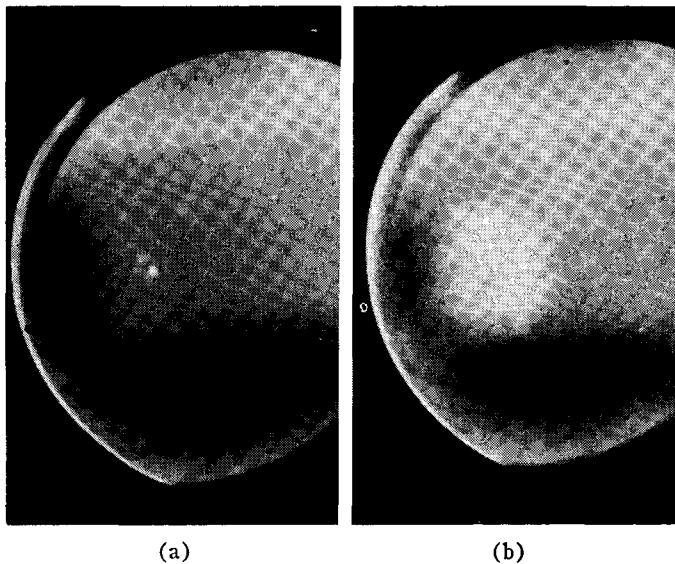

Fig. 4. Oscilloscope displays of the ruby laser (upper trace) and the Raman laser (lower trace) outputs. Sweep rate is 200 μs/division, from left to right.

Fig. 3. Near field patterns of the Raman laser. (a) Input energy is 1.3E_{th}. (b) Input energy is 2.5E_{th}.

Fig. 5. Typical waveforms of the exciting ruby laser spike (the upper trace) and the excited Raman laser spike (the lower trace). Sweep rate is 100 ns/division, from left to right.

cathode has been used to detect the ruby laser spikes.

The threshold ruby laser power to build up the ν_2 Stokes line has been inferred to be 9.5 kW inside the optical resonator from careful observation of sample spikes as shown in Fig. 5. The peak output powers of the ruby and Raman lasers were measured external to the 95 per cent reflecting mirror to be of the order of a few kilowatts and a few hundred watts, respectively.

The biplanar phototube and a traveling-wave oscilloscope have been used to observe the detailed waveform of the Raman laser spikes. An example of the waveform of the Raman laser spikes is shown in Fig. 6. The rise-up and the decaying time constants of the Raman laser spikes have been determined as 1 ns and 6 ns, respectively.

The wiggles observed in the waveform of the Raman laser output are assumed to be the beat note between
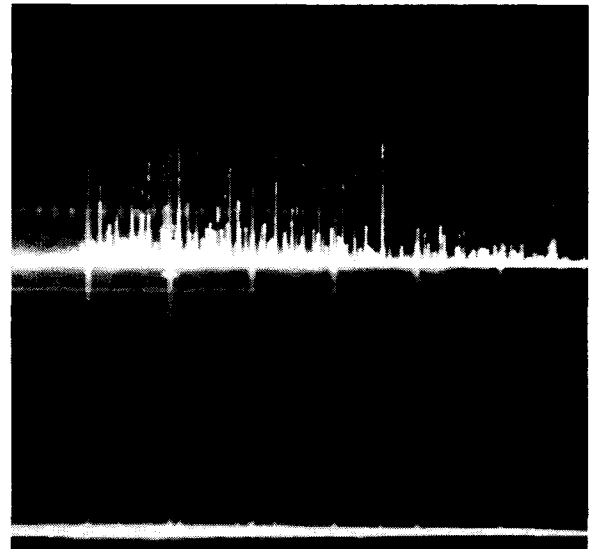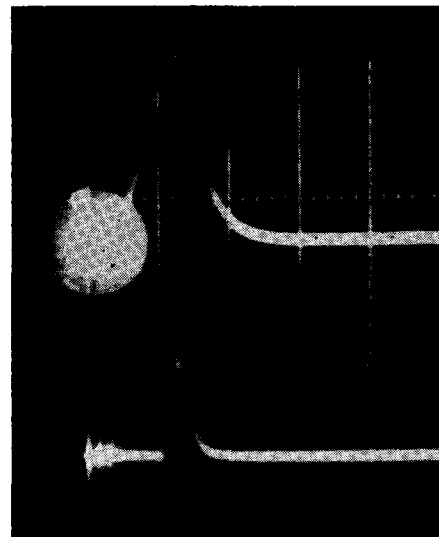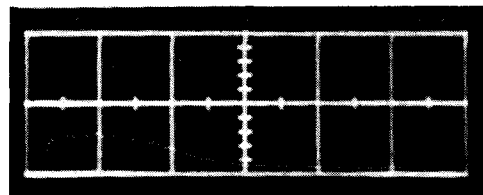

Fig. 6. Detailed waveform of an output spike of the Raman laser. Sweep rate is 10 ns/division, from left to right.

the outputs from the different longitudinal modes. The fundamental frequency of the wiggles is about 400 Mc/s, which agrees with the calculated value of the separation between the nearest resonant frequencies of the two longitudinal modes. The components of the second, the third, and the fourth harmonics of 400 Mc/s have also been found in Fig. 6.

ANALYSIS OF THE EXPERIMENTAL RESULTS

The probability of stimulated Raman scattering is expressed as [3], [4], [6],

$$P = n_m V \eta k n_1 (n_2 + 1) \qquad (1)$$

where n_m is the number of the Raman active molecules per unit volume; V, the volume of the optical resonator; η, the filling factor of the Raman active medium; k, the constant which is determined by the "complex" matrix element [6] of the initial and the final states of the molecule in the Raman scattering and also by the wavelength of the incident light; n_1, the photon density of the ruby laser; and n_2, the photon density of the Raman laser mode which is induced by the ruby laser photons.

In (1), n_1 should be replaced by $\Sigma_i n_{1i} = N_1$, when the photons oscillating in more than one mode are exciting the Raman laser, assuming that k takes the same value in all the exciting ruby laser modes.

The loss of the photons per unit time interval in the optical resonator can be described as $\omega_2(W/Q_2)$, where Q_2 is the quality factor of the optical resonator at the Raman component under consideration and W is the stored radiation energy in the resonator. Therefore, when $n_2 \gg 1$, the rate equation of the Raman laser will be given as

$$\frac{dn_2}{dt} = n_m \eta k N_1 n_2 - \frac{\omega_2 n_2}{Q_2} \qquad (2)$$

From (2) the threshold value of N_1 to build up the oscillation of the Raman laser is given by

$$N_{1\text{th}} = \frac{\omega_2}{n_m \eta k Q_2} \qquad (3)$$

From the experimental results described in the previous section, $N_{1\text{th}}$ has been determined as $5.8 \cdot 10^{13}$. The values on n_m and ω_2 are known and η can be estimated from the lengths of the optical resonator and the Raman cell as 0.43, so that the value of k is determined only if Q_2 is known.

The first term of the right-hand side of (2) is much smaller than the second term, when N_1 is much smaller than $N_{1\text{th}}$. Thus τ, the decay time constant of the tail of the Raman laser spike where N_1 is much smaller than $N_{1\text{th}}$, will be related with Q_2 as $Q_2 = \omega_2 \tau$.

Q_2 has been evaluated as $1.5 \cdot 10^7$, putting $\tau = 6$ ns and $\omega_2 = 2.5 \cdot 10^{15}$ s^{-1}, and we have $k = 1.0 \cdot 10^{-28}$ s^{-1} cm^6 for the ν_2 Stokes line of benzene. The cross section of the incoherent Raman scattering can be calculated from the value of k obtained above. Since the line width of the Stokes component is 4 cm^{-1} when the density of the incident radiation is small [7], [8], the number of modes of the scattered radiation per unit volume is $1.05 \cdot 10^{10}$, the total cross section will be given as $\sigma = 0.46 \times 10^{-28}$ cm^2, taking into account that the scattered ν_2 Stokes line has the same polarization as the incident radiation.

This value of σ for benzene agrees well with the direct measurement of 0.523×10^{-28} cm^2 [7].

However, the extremely high efficiency in generating the stimulated $2\nu_2$, $3\nu_2$, or $4\nu_2$ Stokes lines cannot be understood from the theory described above. The peak output power of the ruby laser should be more than 50 kW to excite up to the $3\nu_2$ Stokes Raman laser, assuming that the $N\nu_2$ Stokes line is excited only by the $(N-1)\nu_2$ Stokes line and the conversion efficiency of 10 per cent in each step. However, the peak output power has been measured to be only of the order of 5 kW.

The intensity distribution in the spectrum of the output of the Raman laser in Fig. 2 shows that the $2\nu_2$ and $3\nu_2$ Stokes lines sometimes have almost the same intensity as the ν_2 Stokes line. This also shows a very high conversion efficiency in stimulating the higher order Stokes lines.

This high efficiency in generating the higher-order Stokes lines should indicate that many multiple mechanisms contribute to build up the higher-order Stokes radiation. For example, the $2\nu_2$ Stokes line is built up not only with Raman scattering of the ν_2 Stokes line, but also by modulation of the ν_2 Stokes line due to the oscillation of the ruby laser and the ν_2 Stokes line [9].

The high efficiency of the higher-order Stokes generation also can be explained by a saturation effect, simply assuming a cascade process in the generation of the stimulated Stokes radiations. When a Raman cell is inserted into the resonator, the peak output of the ruby laser decreases very much. This is because of the saturation effect due to the generation of the Stokes component. The same effect should be expected in the generation of the higher Stokes radiations.

The rate equation of a ruby laser under such conditions will be described as

$$\frac{dn_1}{dt} = b n_p (n_1 + 1) - n_m \eta k n_1 (n_2 + 1) - \frac{\omega_1 n_1}{Q_1} \qquad (4)$$

where n_p is the population difference between the laser energy levels, and b, the constant which gives the transition probability between the laser levels. In the steady state, assuming n_1, $n_2 \gg 1$, we have

$$n_2 = \frac{n_p b}{n_m \eta k} \left(1 - \frac{n_{p\text{th}}}{n_p} \right) \qquad (5)$$

where $n_{p\text{th}} = \omega_1 / b Q_1$ is the threshold value of n_p to build up the ruby laser. Usually $n_p b \gg n_m \eta K$, and n_2 can be very large because $n_p \gg n_{p\text{th}}$ when the stimulated Raman effect is excited by the laser.

On the other hand, the rate equation for the first Stokes radiation will be given as

$$\frac{dn_2}{dt} = n_m \eta k n_1 (n_2 + 1) - n_m \eta k n_2 (n_3 + 1) - \frac{\omega_2 n_2}{Q_2} \qquad (6)$$

where n_3 is the number of the second Stokes photons in

a unit volume. At the stationary condition, assuming $n_2, n_3 \gg 1$, we have

$$n_3 = n_1\left(1 - \frac{n_{1\text{th}}}{n_1}\right) \qquad (7)$$

This equation shows that the second Stokes line can be as intense as the ruby laser line. It seems to disagree with what has been observed, at the first sight. However, the peak output of the Raman laser was measured to be of the same order of magnitude (a few hundred watts inside the resonator) as the threshold laser power to excite the stimulated Raman effect, as previously mentioned. This shows that the previous discussion is qualitatively correct, and probably the larger portion of the ruby laser output is not responsible in stimulating the Raman effect, when the laser is pumped very hard. This may be because of the difference in the cross sections of the optical fibers in which the ruby or the Raman lasers are oscillating. Thus it can be argued that the efficiency of the stimulated Raman effect is much better than its appearance.

Therefore, under a very high exciting power, the incident laser beam, the first-, and the higher-order stimulated Stokes lines should have outputs of about the same order of magnitude if compared relative to their cross-sectional areas.

POSSIBILITY OF FURTHER IMPROVEMENT

It is interesting to consider the possibility of improving the efficiency beyond what we have obtained here by using the results of the present investigation.

It is seen from (3) shows that increases in the values of η and Q_2 will improve the efficiency of the stimulated Raman effect. However, we cannot increase η without interfering with the ruby laser oscillation. The rate equation for the ruby laser which is exciting the Raman laser is

$$\frac{dn_1}{dt} = n_a B n_1 - n_m \eta k n_1 n_2 - \frac{\omega_1 n_1}{Q_1} \qquad (8)$$

and

$$\frac{dn_a}{dt} = N_a - n_a B(n_1 + 1) \qquad (9)$$

where n_a is the number of the effective excited state ions

in unit volume; B, the coefficient for stimulated emission; N_a, the increase of n_a due to the pumping; and Q_1, the quality factor of the optical resonator of the ruby laser.

It is shown in (4) that the higher the value of the second term on the right-hand side, the lower the effective quality factor of the optical resonator; therefore, an increase of η causes a decrease of the ruby laser output.

Thus there is an optimum value of η to get maximum efficiency. On the other hand, Q_2 can be increased as much as possible. In the case of a Raman laser which is excited with a ruby laser, we cannot expect extremely high Q values because of the optical inhomogeneousness of the ruby crystal. If we use a gas laser, it is not unreasonable to assume that $Q_2 = 10^{10}$, and $\eta = 0.2$. Then we get $N_{1\text{th}} = 2 \cdot 10^{11}$ photons/cm^3. This corresponds to the excitation power of about 150 watts, assuming the average cross section of the optical beam to be 0.1 cm^2. It may be possible to construct a CW Ar gas laser, the radiation power of which is more than 150 watts inside the optical resonator. The previous discussion shows the possibility of exciting a stimulated Raman laser by inserting an optically homogeneous Raman cell inside the resonator of a high-gain Ar gas laser. In other words, the efficiency of the stimulated Raman laser can be improved more than 30 times beyond the system reported in this paper.

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