PRINCIPLES OF RAMAN LASERS AND PARAMETRIC AMPLIFIERS

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With the advent of powerful lasers at selected frequencies, it has now become possible to generate new frequencies by stimulating Raman scattered waves of these light sources. In a medium with high second-order non-linear optical susceptibility, new frequencies are also generated by parametric conversion. Principles of stimulated Raman scattering and the three-wave parametric interaction in a non-linear medium are presented and compared with the physical process involved in the operation of a conventional laser. As two recent examples of tunable devices based on these principles, the case of lithium niobate ($LiNbO_3$), where a polariton mode is excited, and the case of n-type InSb in a magnetic field, where an electron is excited to an empty spin-level, are discussed. The device using $LiNbO_3$ crystal is capable of generating tunable infra-red sources from 50 μ to 250 μ whereas InSb device can generate tunable sources in the range 6–13 μ.

INTRODUCTION

For many applications of intense light beams in both pure and applied physics work, one needs a monochromatic radiation source of frequency different than that available from conventional fixed frequency laser sources. Frequency conversion is one of the important features which we plan to discuss in this article. The aim is to understand how one uses different physical processes to obtain intense coherent light sources in the optical and the infra-red frequency regions.

Basically there are three relevant processes to generate intense light sources:

(i) stimulated emission from an upper level to a lower atomic level (conventional laser),
(ii) stimulated Raman scattering (Raman laser), and
(iii) three-wave parametric interaction in a non-linear medium (parametric oscillator).

In order to bring out the differences in working of devices based on these processes, we will first discuss the physical principles involved in their operations. As examples of Raman lasers and parametric oscillators, we will then describe two recent devices obtained by using n-type InSb and $LiNbO_3$.

CONVENTIONAL LASERS

It is well known that the operation of a conventional laser (Schawlow and Townes 1958) depends upon the utilization of the phenomenon of

stimulated emission from a population inverted excited atomic level. The attainment of the population inversion is achieved by means of an external agency, usually referred to as 'pump' (Fig. 1). According to quantum theory

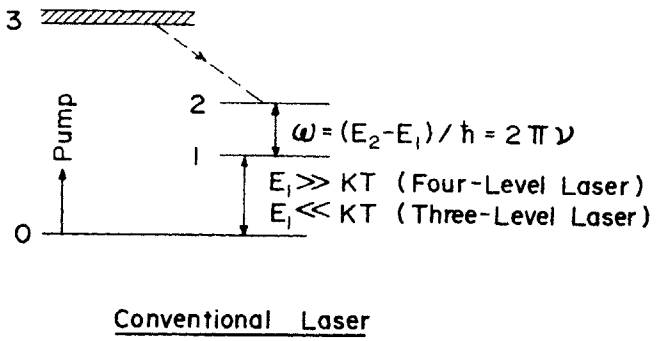

FIG. 1. Atomic energy level diagram showing laser transition between levels 1 and 2.

of radiation, since photons are Bose particles, the rate of emission of photons in a single mode is proportional to $(n_\lambda+1)$ and the rate of the absorption to n_λ, where n_λ is the number of photons in the mode. The stimulated emission refers to n_λ in $(n_\lambda+1)$ whereas the spontaneous emission refers to 1 in $(n_\lambda+1)$. Since spontaneous emission takes place for all the modes for which energy conservation is satisfied, spontaneous transition rate is obtained by summing over all the modes within the natural line width of the atomic levels. If $\Delta\nu$ is the width of the line, assumed to be Lorentzian, there are $p(\nu) = 8\pi\nu^2\Delta\nu V/c^3$ modes (in volume V) for the spontaneous transition. Therefore, induced emission rate for given mode λ is obtained by dividing the spontaneous transition rate $(1/\tau_{\text{spont}})$ by $p(\nu)$ and multiplying it by n_λ. If the atomic medium with an inverted population is present within an optical resonator (Schawlow and Townes 1958), it would amplify the mode energy. If the amplification is sufficient to compensate for whatever loss mechanism (absorption, scattering, etc.) may be present, the energy density will increase in time, and an oscillation near the resonant frequency of the resonator will be established.

In a simple treatment like this, the rate of increase of n_λ is given by

$$\frac{dn_\lambda}{dt} = n_\lambda\left[\frac{(N_2-N_1)c^3}{\tau_{\text{spont}} V\, 8\pi\nu^2\Delta\nu}\right] - \frac{n_\lambda}{\tau_\lambda} \quad .. \quad .. \quad .. \quad (1)$$

where $\tau_\lambda = Q/\omega$ is the decay time, due to losses in the resonator, and (N_2-N_1) is the population difference between the upper and lower atomic levels. If $N_2 > N_1$, the steady state is reached if the gain associated with the first term in eqn. (1) is compensated by the loss, i.e.

$$g \equiv \frac{(N_2-N_1)c^3}{\tau_{\text{spont}} V\, 8\pi\nu^2\Delta\nu} = \frac{1}{\tau_\lambda} \quad .. \quad .. \quad .. \quad .. \quad (2)$$

or

$$(N_2-N_1) = \frac{\tau_{\text{spont}} V 8\pi\nu^2 \Delta\nu}{\tau_\lambda c^3}. \qquad \ldots \quad \ldots \quad \ldots \quad (3)$$

It is to be noted that for amplification (N_2-N_1) has to be large, τ_{spont} small, and the natural line width $\Delta\nu$ small.

The excitation (pumping) mechanism to obtain considerable population difference (N_2-N_1) differs considerably in various types of lasers. In solid state lasers, like Ruby or YAG : Nd^{3+}, it is achieved through optical pumping. However, in gas lasers it is done through vibrational energy transfer, resonance energy exchange or electron collision excitations. In dye lasers one uses another laser or specially designed flashlamps for pumping. Organic dye lasers are of special importance because these can be tuned over a wide frequency region in the visible and near infra-red by changing dye concentrations.

Raman Lasers

Next let us consider stimulated Raman scattering (Bloembergen 1967). By Raman scattering we mean inelastic light scattering in general. In any inelastic process an incident light frequency ω_i is scattered to frequency ω_s. For the Stokes component, which we would consider here, $\omega_s < \omega_i$, and the system is left in an excited state. In solids, the optical-phonon-Raman scattering is due to the excitation of an optical phonon whereas the acoustic-phonon-Raman scattering or the Brillouin scattering involves the excitation of an acoustic phonon. In these processes the electronic state of the sample remains the same. However, consider the case of a n-type semi-conductor (Jha 1969). The charged carriers can be excited either as single particles or collectively by creating plasmons with frequency $\omega_p \simeq (4\pi ne^2/m^*)^{\frac{1}{2}}$. Because of continuous energy levels, the single-particle electronic excitations are quite weak in the long wavelength limit, and most of the scattering in this case is due to the excitation of plasmons. However, if one applies a strong d.c. magnetic field H, the motion of the carriers perpendicular to the field is quantized into Landau levels with separation in frequency approximately equal to eH/m^*c. Because of the spin-moment, these levels are further split due to the magnetic field with frequency separation $g\beta H/\hbar = geH/2mc$, where g is the effective g-factor for the carriers (Fig. 2). At low temperatures and for carrier concentrations of the order of 10^{16}–10^{17} cm^{-3}, only the lowest level or only a few of the lower ones are occupied by the carriers. In a solid, because of the non-parabolicity of the band-energy, light can excite electrons between different Landau-levels, and because of the spin-orbit interaction it can excite electrons from one spin-level to another (Jha and Rao 1969). The excitation due to electronic transitions between Landau-levels ($\Delta l = 1$ or $\Delta l = 2$, $\Delta S = 0$) is called Landau-Raman scattering, whereas the excitation

between different spin-levels ($\Delta S = 1$, $\Delta l = 0$) is called spin-flip-Raman seattering.

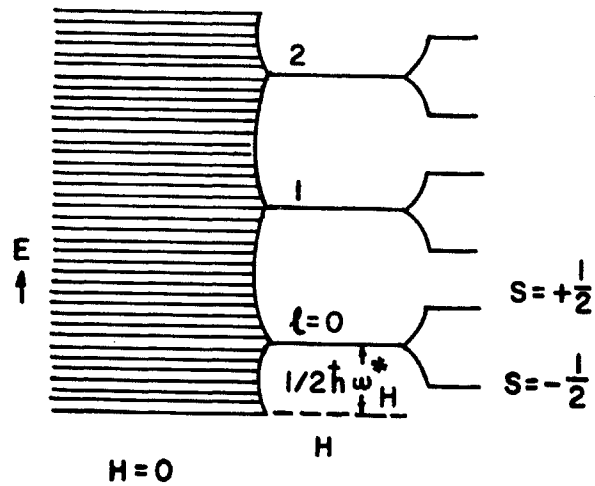

FIG. 2. Carrier energy levels (spin $\frac{1}{2}$) in a magnetic field H.

The transition probability for all these excitations, whether electronic or ionic, is proportional to $n_i(n_s+1)$, where n_i and n_s are number of photons in the incident and the scattered mode respectively. The number 1 in (n_s+1) gives the well-known spontaneous scattering, being studied for the case of ionic excitations for more than four decades, whereas n_s in (n_s+1) refers to the stimulated scattering. If $(d\sigma/d\Omega)_{\text{spont}}$ is the scattering cross-section (per unit solid angle) for a given polarization,

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{stimulated}} = n_s\left(\frac{d\sigma}{d\Omega}\right)_{\text{spont}}\left(\frac{c^3}{V\nu_s^2\Delta\nu}\right) \quad .. \quad .. \quad (4)$$

where we have divided the spontaneous cross-section by the number of scattering photon modes per unit solid angle (for one polarization) within the linewidth $\Delta\nu$ of the spontaneous line, and multiplied by n_s. Thus the rate of increase of the scattered photon at a finite temperature T is given by

$$\frac{dn_s}{dt}+\frac{n_s}{\tau_s} = \frac{n_s n_i c}{V}\left(\frac{1}{V}\frac{d\sigma}{d\Omega}\right)_{\text{spont}}\frac{c^3}{\nu_s^2\Delta\nu}[1-\exp\{-\hbar(\omega_i-\omega_s)/kT\}] \quad .. \quad (5)$$

or

$$n_s \sim e^{\left(g-\frac{1}{\tau_s}\right)t} \quad .. \quad .. \quad .. \quad .. \quad (6)$$

where

$$g = \frac{n_i c}{V}\left(\frac{1}{V}\frac{d\sigma}{d\Omega}\right)_{\text{spont}}\frac{c^3}{\nu_s^2\Delta\nu}[1-\exp\{\hbar(\omega_i-\omega_s)/kT\}] \quad .. \quad .. \quad (7)$$

and where τ_s is the lifetime of the scattered photon in the medium. If we assume that end faces of the sample are perfectly reflecting so that losses are

mostly due to bulk absorption, $\tau_s \simeq \mu/c\alpha_s$, where μ is the refractive index of the sample and α_s is the absorption coefficient in cm^{-1}. The threshold for stimulated scattering is obtained by equating the gain coefficient g to $1/\tau_s$. The threshold power per cm^2 for the incident beam is, therefore, given by

$$(P_{in})_{thres} = \frac{n_i c}{V}\hbar\omega_i = \frac{\hbar\omega_i}{\tau_s}\left[\left(\frac{1}{V}\frac{d\sigma}{d\Omega}\right)_{spont}\frac{c^3}{\nu_s^2\Delta\nu}[1-\exp\{-\hbar(\omega_i-\omega_s)/kT\}]\right]^{-1} \quad (8)$$

It has to be noted that for low threshold $(d\sigma/d\Omega)_{sp}$ has to be large, and the linewidth $\Delta\nu$ of the spontaneous line small. One must choose only those lines which are sharp and intense. Although, in principle, each spontaneous light-scattering process has a stimulated counterpart, not all lines can be stimulated by using incident powers available from conventional lasers. Also when one uses intense incident beams, several other non-linear processes like self-focusing due to intensity-dependent refractive index, multi-photon absorption, etc., start competing with the stimulated light scattering of various types. To be consistent one has also to consider then the anti-Stokes line in the analysis (Bloembergen 1967).

It has to be emphasized that no population inversion is involved in the stimulated Raman scattering. However, for most cases, the incident threshold power comes out to be in the region of kilowatts/cm^2, and one has to use another laser as the incident beam. The incident light frequency can be arbitrary except to the limitation that it should not be absorbed considerably by the sample. But $(d\sigma/d\Omega)_{spont}$ can be increased appreciably if one takes advantage of the 'resonant' scattering, i.e. if the incident frequency is such that it connects a real intermediate state, scattering being a second-order process (Fig. 3).

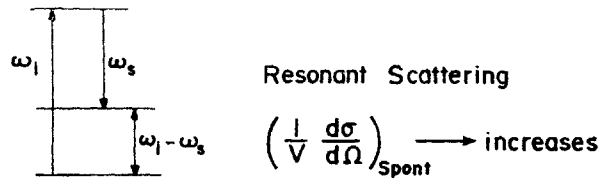

FIG. 3. Resonant Raman scattering process. ω_i and ω_s are incident and scattered frequencies respectively.

PARAMETRIC AMPLIFIERS AND OSCILLATORS

Let us now discuss the case of parametric amplifiers. In crystals lacking inversion symmetry, electromagnetic waves of different frequencies are coupled because there is a finite second-order non-linear polarization induced in the solid (Armstrong *et al.* 1962). Phenomenologically, the non-linear polarization at frequency ω is given by

$$P^{NL}(\omega) = \sum_{\omega'} \overleftrightarrow{\chi}^{NL}(\omega', \omega-\omega') : \vec{E}(\omega')\vec{E}(\omega-\omega') \quad .. \quad .. \quad (9)$$

which enters in Maxwell's equations as

$$\nabla^2\vec{E}(\omega)+\frac{\mu^2(\omega)\omega^2}{c^2}\vec{E}(\omega)+\frac{4\pi i\omega}{c^2}\sigma_\omega\vec{E}(\omega) = -\frac{4\pi\omega^2}{c^2}\vec{P}^{NL}(\omega), \quad .. \quad (10)$$

where $\mu^2(\omega)$ is the real part of the linear dielectric function $\epsilon(\omega)$, and the conductivity $\sigma(\omega)$ is related to the imaginary part of the linear dielectric function: $\sigma(\omega) = \omega\, Im\,\epsilon(\omega)/4\pi$. Note that we have neglected, for simplicity, dispersive terms in Maxwell's equations.

Let us consider only three modes, the frequencies ω_1, ω_2 and ω_3, such that

$$\omega_3 = \omega_1+\omega_2. \quad .. \quad .. \quad .. \quad .. \quad (11)$$

Also let

$$E_i = \tfrac{1}{2}[E(\omega_i, z)e^{-i\omega t}+E^*(\omega_i, z)e^{i\omega t}],$$

$$E(\omega_i, z) \equiv E_i z) = A_i(z)e^{ik_i z},\ K_i = \frac{\omega_i\mu_i}{c}, \quad .. \quad .. \quad (12)$$

specializing the problem to one dimension by assuming propagation along z.

If there was no interaction between the waves, except for the exponential damping because of the linear loss term, we would have independent plane waves as solutions of Maxwell's equations satisfied by E_1, E_2 and E_3. However, because of the non-linear interaction the variation of $A_i(z)$ is no longer governed by the loss term alone. In the slowly varying envelope approximation (SVEA) (Armstrong *et al.* 1962, Yariv 1967) where one still assumes $A_i(z)$ to be a slowly varying function of z, Maxwell's equations reduce (Armstrong *et al.* 1970) to:

$$\frac{\partial A_3}{\partial z} = i\frac{2\pi\omega_3}{c\mu_3}\chi^{NL}A_1A_2e^{-i(K_3-K_1-K_2)z}-\alpha_3A_3 \quad .. \quad .. \quad (13)$$

$$\frac{\partial A_2}{\partial z} = i\frac{2\pi\omega_2}{c\mu_2}\chi^{NL}A_3A_1{}^*e^{i(K_3-K_1-K_2)z}-\alpha_2A_2 \quad .. \quad .. \quad (14)$$

$$\frac{\partial A_1}{\partial z} = i\frac{2\pi\omega_1}{c\mu_1}\chi^{NL}A_3A_2{}^*e^{i(K_3-K_1-K_2)z}-\alpha_1A_1 \quad .. \quad .. \quad (15)$$

where $\alpha_i = 2\pi\sigma_i/c\mu_i$ is the absorption coefficient in cm^{-1}.

In order to understand the parametric conversion process let us first assume that

$$\Delta K = K_3-K_1-K_2 = 0 \text{ (phase matching)}$$

and

$$\alpha_i = 0 \text{ (lossless medium)}.$$

To consider the process of amplification of a weak signal at ω_1 by a pump wave at ω_3, in the first approximation one can assume $A_3(z)$ to be constant and real. Then

$$\frac{\partial A_1}{\partial z} \approx i\frac{G}{2}\left(\frac{\omega_1\mu_2}{\omega_2\mu_1}\right)^{\frac{1}{2}}A_2^* \quad .. \quad .. \quad .. \quad .. \quad (16)$$

$$\frac{\partial A_2^*}{\partial z} \approx -i\frac{G}{2}\left(\frac{\omega_2\mu_1}{\omega_1\mu_2}\right)^{\frac{1}{2}}A_1. \quad .. \quad .. \quad .. \quad (17)$$

For $A_2(0) = 0$, we therefore obtain

$$|A_1|^2 = |A_1(0)|^2 \cosh^2 \frac{G}{2} z \quad .. \quad .. \quad .. \quad .. \quad (18)$$

$$|A_2|^2 = |A_1(0)|^2 \left(\frac{\omega_2 \mu_1}{\omega_1 \mu_2}\right) \sinh^2 \frac{G}{2} z, \quad .. \quad .. \quad .. \quad (19)$$

where G, the gain per unit length, is given by

$$G \equiv \frac{4\pi}{c} \left(\frac{\omega_1 \omega_2}{\mu_1 \mu_2}\right)^{\frac{1}{2}} \chi^{NL} A_3(0). \quad .. \quad .. \quad .. \quad .. \quad (20)$$

For $\Delta K \neq 0$, eqns. (16) and (17) are replaced by

$$\frac{\partial A_1}{\partial z} = i \frac{G}{2} \left(\frac{\omega_1 \mu_2}{\omega_2 \mu_1}\right)^{\frac{1}{2}} A_2^* e^{i \Delta K z} \quad .. \quad .. \quad .. \quad .. \quad (21)$$

$$\frac{\partial A_2^*}{\partial z} \approx -i \frac{G}{2} \left(\frac{\omega_2 \mu_1}{\omega_1 \mu_2}\right)^{\frac{1}{2}} A_1 e^{-i \Delta K z} \quad .. \quad .. \quad .. \quad (22)$$

which, with $A_2(0) = 0$, lead to

$$A_i(z) = A_1(0) e^{\frac{i \Delta K z}{2}} \left[\cosh \frac{gz}{2} - \frac{i \Delta K}{g} \sinh \frac{gz}{2}\right] \quad .. \quad .. \quad (23)$$

where

$$g = \sqrt{G^2 - (\Delta K)^2}. \quad .. \quad .. \quad .. \quad (24)$$

Thus for amplification $(\Delta K)^2$ must be smaller than G^2. The gain is maximum if $\Delta K = 0$, i.e. if there is perfect momentum conservation. In undepleted pump approximation the effect of absorption can also be calculated by adding these terms in eqns. (21) and (22). However, it is obvious that for significant power conversion α_i has to be small compared to the non-linear interaction length G^{-1}. For $LiNbO_3$, $\chi^{NL} \sim 10^{-8}$ esu, which implies that $G^{-1} \simeq 2$ cm for $|A_3| = 150$ esu ($P_3 \sim 5 \times 10^6$ watts/cm^2).

The exact solution for the coupled wave equations (13)–(15) (with $\alpha_i = 0$), when the non-depleted pump approximation is not used, is given by Armstrong 1962 and is sketched here in Fig. 4.

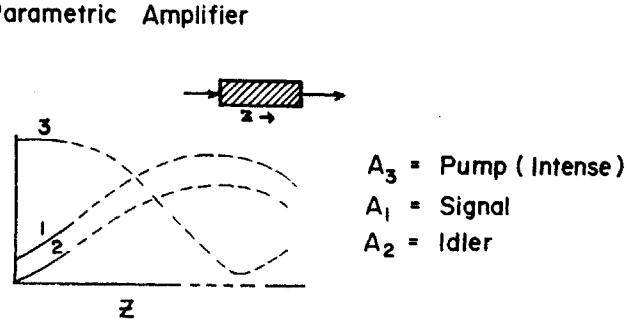

FIG. 4. Parametric amplification of signal at frequency ω_i as a function of z.

In order to describe the interaction inside an optical resonator in the time domain, we must consider the variation of the field as seen by an observer travelling with velocity c/μ_i of the optical waves. Thus for the perfect phase-matching case we obtain

$$\frac{\partial A_3}{\partial t} = i\frac{2\pi\omega_3}{\mu_3{}^2}\chi^{NL}A_1A_2 - \frac{A_3}{\tau_3} \quad .. \quad .. \quad .. \quad .. \quad (25)$$

$$\frac{\partial A_2{}^*}{\partial t} = -i\frac{2\pi\omega_2}{\mu_2{}^2}\chi^{NL}A_3{}^*A_1 - \frac{A_2{}^*}{\tau_2} \quad .. \quad .. \quad .. \quad (26)$$

$$\frac{\partial A_1}{\partial t} = i\frac{2\pi\omega_1}{\mu_1{}^2}\chi^{NL}A_3A_2{}^* - \frac{A_1}{\tau_1} \quad .. \quad .. \quad .. \quad (27)$$

where $\tau_i = \mu_i/c\alpha_i$. To obtain the threshold for steady state oscillation, with A_3 as input, we put $\partial A_2{}^*/\partial t = \partial A/\partial t = 0$, and obtain

$$|A_3|^2_{\text{thres.}} = \frac{\mu_1{}^2\mu_2{}^2}{4\pi^2\omega_1\omega_2\tau_1\tau_2(\chi^{NL})^2}$$

which is equivalent to the condition

$$G = \sqrt{\alpha_1\alpha_2} \text{ (threshold)} \quad .. \quad .. \quad .. \quad .. \quad (28)$$

where G is given by eqn. (20). If $|A_3|^2$ is greater than the threshold, we have a direct method of converting input frequency at ω_3 to output frequencies ω_1 and ω_2.

The set of eqns. (13)–(15) can also be used to analyse parametric up-conversion in which ω_1 (pump) and ω_2 are initially present, and ω_3 is generated. The second harmonic generation, with $\omega_1 = \omega_2$, is a special case of this process. In this special case, where $\omega_3 = 2\omega_1$, we have

$$\frac{\partial A_3}{\partial z} = i\frac{2\pi}{c}\frac{\omega_3}{\mu_3}\chi^{NL}A_1{}^2e^{-i\Delta Kz} \quad .. \quad .. \quad .. \quad (29)$$

so that

$$|A_3|^2 = \frac{4\pi^2\omega_3{}^2}{c^2\mu_3{}^2}(\chi^{NL})^2(A_1{}^2)^2z^2\frac{\sin^2(\Delta Kz/2)}{(\Delta Kz/2)^2} \quad .. \quad .. \quad (30)$$

which is maximum for $\Delta K = 0$. If $\Delta K \neq 0$, the second harmonic power goes through a succession of zeroes and maxima as a function of z, with separation $l_{\text{coh}} = \pi/\Delta K$ (coherence length).

Two Recent Devices

We conclude this paper by reporting two recent developments in which ideas relating to stimulated Raman scattering and parametric interactions have been used in a most dramatic manner. The first one uses spin-flip Raman scattering of a CO_2 (10·6 μ) or a CO (5·3 μ) laser from carriers in InSb to obtain a tunable Raman laser in the infra-red. Since the frequency of the scattering light $\omega_s = \omega_i - g\beta H/\hbar$, it can be easily tuned by varying the magnetic field. This process was first suggested (Jha and Rao 1969) in 1968–69.

Patel *et al.* (1970) was in fact able to stimulate the spin-flip line in InSb by using a CO_2 laser. The Raman laser was tunable from 10·9 μ to 13·0 μ by varying H from 16 kg to 100 kg. With incident power of 1·5 kW inside the crystal, output power of the order of 30–100 w was obtained. Taking advantage of resonant scattering by using a CO laser, Mooradian (*in press*) of Lincoln Labs. was later able to decrease the threshold power to less than 0·1 watt/cm^2 and obtain a CW Raman laser in the infra-red.

The second device of Yarborough *et al.* (1969) consists of a lithium niobate ($LiNbO_3$) crystal placed in front of a Q-switched ruby laser. The input pump photon from the ruby laser excites an infra-red active 248 cm^{-1} optical phonon (polariton, to be exact) and emits a photon in the visible at the Stokes frequency. The lattice vibration, however, generates radiation at the infra-red frequency since it is infra-red active. The device is thus based on the coupled mechanism of the stimulated Raman scattering and parametric amplification. Rotation of the crystal changes the frequencies involved because of the angular dependence of the dispersive properties of the crystal, the polariton mode being highly dispersive in the small wavenumber region. In the infra-red the device is tunable continuously from 50 μ to 250 μ with powers of the order of 10–100 watts. In the visible it is tunable over about 100 Å with a conversion efficiency of about 50 per cent from a 1 mw ruby laser.

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