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Physics of Raman Lasers*

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The physics of Raman lasers is described in simple terms. A simplified gain formula is presented for the threshold of Raman-laser action. The features of stimulated Raman scattering are compared with those of spontaneous Raman process. Finally, a review of the various cavities used for Raman lasers is presented.

INTRODUCTION

RECENTLY the study of Raman lasers has attracted the attention of both pure and applied scientists. As a topic of basic science, the study of the interactions involved in Raman lasers is fascinating. On the other hand, the use of Raman shifting techniques provides a knob for a gross tuning of the laser frequency to make possible new laser applications. For example, this technique can be used to match the resonance frequency of an atmospheric constituent in an experiment where the laser beam is used as a probe.

The exact details of the interactions, especially in the focused beam are still being unraveled; but the main feature is largely understood. The purpose of this article is to present a simple picture of the phenomenon. The Raman laser utilizes the principle of stimulated Raman scattering. Just like fluorescence emission, scattering can be spontaneous or stimulated. Stimulated scattering exhibits the usual laserlike properties; therefore, the Raman laser can be easily understood in comparison with the ordinary laser.

The spontaneous Raman scattering consists of the scattering of a photon by an atom or a molecule (or a crystal) with a change in frequency. Figure 1 represents the vibrational Raman scattering by a molecule. The vibrational states of the ground electronic state are designated by m and n, while the upper electronic state is designated by r. To observe vibrational Raman effect, a light of frequency ν (where ν is smaller than ν_{rm} but larger than ν_{mn}) is made to excite the molecule to a virtual state i; if the electron returns to n, a Stokes photon of frequency $\nu_R=\nu-\nu_{mn}$ is generated. Similarly, there will be anti-Stokes lines if the molecule is excited from n and it returns to m.

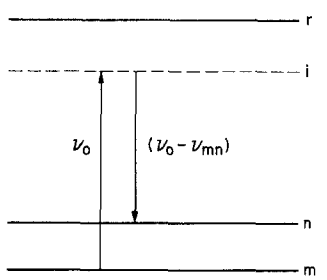

FIG. 1. The energy-level scheme and the transitions in vibrational Raman effect in a molecule.

The above spontaneous Raman scattering has a very low probability (of the order of 10^{-6}); but, as mentioned earlier, in addition to the spontaneous scattering there is another process, viz., the stimulated scattering. In this process, the presence of the scattered photon of frequency ν_R induces an additional probability for scattering of an incident photon of frequency ν as a photon of frequency ν_R. This probability term is dependent on the intensity of both the primary and the scattered radiation and can therefore be controlled just as in ordinary lasers.

I. GAIN FORMULA NEAR THRESHOLD

Following Ehrenfest and Einstein,[1] the probability of the Raman emission per molecule can be written as

$$I_L(A_{mn}+B_{mn}I_R), \tag{1}$$

where A_{mn} and B_{mn} are the probabilities of the spontaneous and stimulated scattering, respectively, and I_L and I_R are the intensities of the exciting laser and the stimulating Raman radiations, respectively, A_{mn} and B_{mn} are related[2] by

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[1] P. Ehrenfest and A. Einstein, Z. Physik **19**, 301 (1923).
[2] A. Mitchell and M. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, Cambridge, England, 1961), p. 94.

the equation

$$A_{mn}=(2h\nu_R^3/c^2)B_{mn}. \quad (2)$$

Therefore, the intensity due to Raman scattering is given by

$$I_L A_{mn} h\nu_R + I_L I_R B_{mn} h\nu_R. \quad (3)$$

An expression for A_{mn} can be obtained in terms of the polarizability matrix element, since the intensity of the spontaneous Raman scattering is given[3] by

$$dI=\frac{2^7\pi^5}{3^2c^4}\nu_R^4\alpha_{mn}^2 I_L \quad (4)$$

where $\alpha_{mn}^2=\sum_{\rho,\sigma}|(\alpha_{\rho\sigma})_{mn}|^2$ is the square of the $\rho\sigma$th matrix element[4] of the polarizability tensor for transition $m\rightarrow n$. Values of α_{mn} can be found[5] experimentally from the measurement of Raman intensities or scattering cross section. By comparing Eq. (4) with the spontaneous scattering part of Eq. (3), one obtains a value for A_{mn}:

$$A_{mn}=(2^7\pi^5\nu_R^3/3^2hc^4)\alpha_{mn}^2. \quad (5)$$

Substituting the above value of A_{mn} in Eq. (2), an expression for B_{mn} is obtained:

$$B_{mn}=(2^6\pi^5/3^2h^2c^2)\alpha_{mn}^2. \quad (6)$$

This result can also be reached directly using the expression for the electric moment of transition. In a Raman laser at higher intensities, the term $A_{mn}I_L$ due to spontaneous scattering can be neglected as compared to the term $B_{mn}I_LI_R$ due to stimulated scattering. Therefore, the Raman intensity due to stimulated scattering is given by

$$d(I_R\delta\nu_R)=I_LI_RB_{mn}h\nu_R. \quad (7)$$

If N is the number of molecules per cubic centimeter, the number of molecules in a volume of unit area and length dx is Ndx; therefore, the increase in intensity in traversing a length dx is given by

$$dI_R=NI_L(I_R)/(\Delta\nu_R)B_{mn}h\nu_R dx, \quad (8)$$

remembering that the stimulative intensity I_R is generated by spontaneous scattering in a bandwidth $\Delta\nu_R$, the linewidth for spontaneous Raman scattering. Substituting the value of B_{mn}, we get

$$dI_R/I_R=(2^6\pi^5N\nu_R/3^2hc^2\Delta\nu_R)\alpha_{mn}^2 I_L dx. \quad (9)$$

Integrating over a distance l and taking the initial value of I_R to be ${}^0I_R$

$$\ln(I_R/{}^0I_R)=(2^6\pi^5N\nu_R/3^2hc^2\Delta\nu_R)\alpha_{mn}^2 I_L\cdot l. \quad (10)$$

Therefore, the gain is given by

$$G=(I_R/{}^0I_R)=\exp[(2^6\pi^5N\nu_R/3^2hc^2\Delta\nu_R)\alpha_{mn}^2\cdot I_L\cdot l]. \quad (11)$$

The threshold for Raman-laser action is reached when the above gain overcomes the losses in the cavity. This gain formula determining the threshold would be true for gases where the refractive index is very close to unity. Some corrections are needed for application to condensed phases.

II. STIMULATED VERSUS SPONTANEOUS RAMAN SCATTERING

The above analysis shows that any material which is efficient in spontaneous Raman scattering may also be efficient for stimulated Raman scattering. The subject of spontaneous Raman scattering has been extensively studied since its discovery in 1928, and therefore, a wealth of information is available, especially regarding wavelengths.[6] However, one important item of information is lacking; this is the width of the scattered line. As shown in Eq. (11), the intensity of the Raman laser is inversely proportional to the linewidth of the spontaneous scattering. Therefore, the lower the linewidth, the lower is the threshold for the Raman laser.

Usually, the spontaneous Raman spectrum consists of a few lines, some of them often having equal intensities. In such a case, the line having the highest intensity with smallest linewidth will be the first to appear in the stimulated

[3] J. Behringer and J. Brandmüller, Z. Electrochem. 60 643 (1956).

[4] The explicit form of the $\rho\sigma$th component of the matrix element α_{mn} is given by the quantum-mechanical dispersion theory to be

$$(\alpha_{\rho\sigma})_{mn}=\frac{1}{h}\sum_r\left[\frac{(\mathbf{M}_\rho)_{rn}(\mathbf{M}_\sigma)_{mr}}{\nu_{rm}-\nu_0}+\frac{(\mathbf{M}_\rho)_{mr}(\mathbf{M}_\sigma)_{rn}}{\nu_{rn}+\nu_0}\right],$$

where $(\mathbf{M}_\rho)_{mr}=\int\psi_r^*\mathrm{m}_\rho\psi_m d\tau$, and so on, with ψ_r and ψ_m as the vibronic wavefunctions and m_ρ as the ρth component of the electric moment operator.

[5] T. Yoshino and H. T. Bernstein, J. Mol. Spectry. 2, 241 (1958).

[6] K. W. F. Kohlrausch, *Der Smekal–Raman Effect* (Julius Springer-Verlag, Berlin, 1931); J. H. Hibben, *The Raman Effect and Its Chemical Application* (Rheinhold Publ. Corp., New York, 1939).

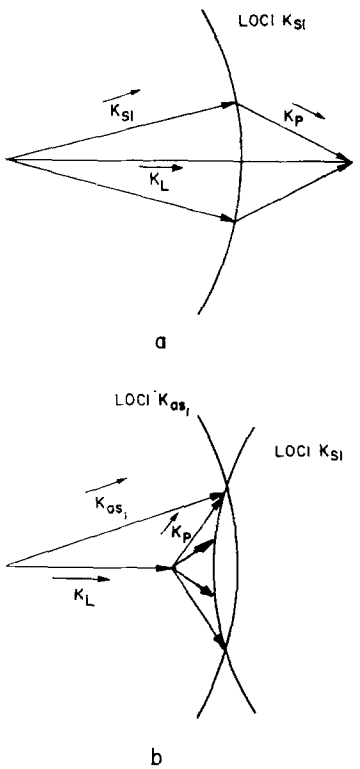

FIG. 2. Momentum-matching conditions for (a) first Stokes and (b) first anti-Stokes Raman processes.

Raman spectrum. It can be shown easily that the totally symmetric vibrations of the molecules usually have the highest scattering cross section with smallest linewidth. In the absence of the data on linewidths, the above criterion can, therefore, be taken as a guide in selection of wavelengths.

The stimulated Raman scattering, however, is distinguished by several distinctly different features as compared to the normal Raman scattering. These are the following:

(1) The spontaneous Raman spectrum consists of many lines corresponding to the various allowed vibrational modes. The stimulated Raman spectrum consists of lines corresponding to one or two vibrational modes. This is due to the fact that when the threshold of the Raman laser action is reached corresponding to one vibrational mode, I_R for that mode starts to increase while I_L decreases; thus, the probability of scattering by that mode becomes increasingly higher compared to the other modes. However, with increasing power, the intensity of the first Raman line can be so high that it can generate its own Raman line, and this can result in a succession of lines of higher order. Thus, the Raman-laser spectrum consists of several orders of Stokes and anti-Stokes lines of one or two vibrational modes.

(2) The other distinguishing feature is that while the spontaneous Raman scattering is incoherent, the stimulated Raman scattering becomes coherent as a natural consequence of the phenomenon of stimulation.

(3) A strikingly different feature of the stimulated Raman scattering is its directionality[7] of emission. This comes as a result of the momentum-matching condition in a dispersive medium. Both energy and momentum are conserved in an interaction between radiation and matter. If a single photon is scattered by a single molecule, the molecule recoils suitably to conserve the momentum. But when a coherent beam interacts with the molecules constituting a medium, the difference of momentum is taken up by the optical phonon.

Therefore, the proper description of the Raman-laser interaction should be in terms of the interaction of coherent plane waves, although so far we have considered interactions of individual particles for the ease of understanding. The first Stokes radiation comes diffusely in a cone around the exciting laser beam. The anti-Stokes radiation comes in sharply defined cones of radiation with increasing apex angles for the higher orders. This can be easily understood in terms of the wave vectors, since the various waves in the Raman laser can be considered as coherent plane waves and, therefore, each can be represented by a wave vector $\mathbf{k}$. The condition for the production of first Stokes radiation is

$$\mathbf{k}_L = \mathbf{k}_{S1} + \mathbf{k}_p, \tag{12}$$

where $\mathbf{k}_L$, $\mathbf{k}_{S1}$, and $\mathbf{k}_p$ are the wave vectors for the exciting laser, the Raman laser, and the optical phonon. Since the magnitude of k_{S1} is fixed by the condition of the conservation of energy, the above relation is satisfied at all points over the circle of radius k_{S1} as shown in Fig. 2(a). But when the Stokes beam gets away from the exciting laser beam, the overlap between the

[7] E. Garmire, F. Pandarese, and C. H. Townes, Phys. Rev. Letters 11, 160 (1963); H. T. Zeiger, P. E. Tannenwald, S. Kern, and R. Herendeen, *ibid.*, p. 419; R. Chiao and B. P. Stoicheff, *ibid.*, p. 290.

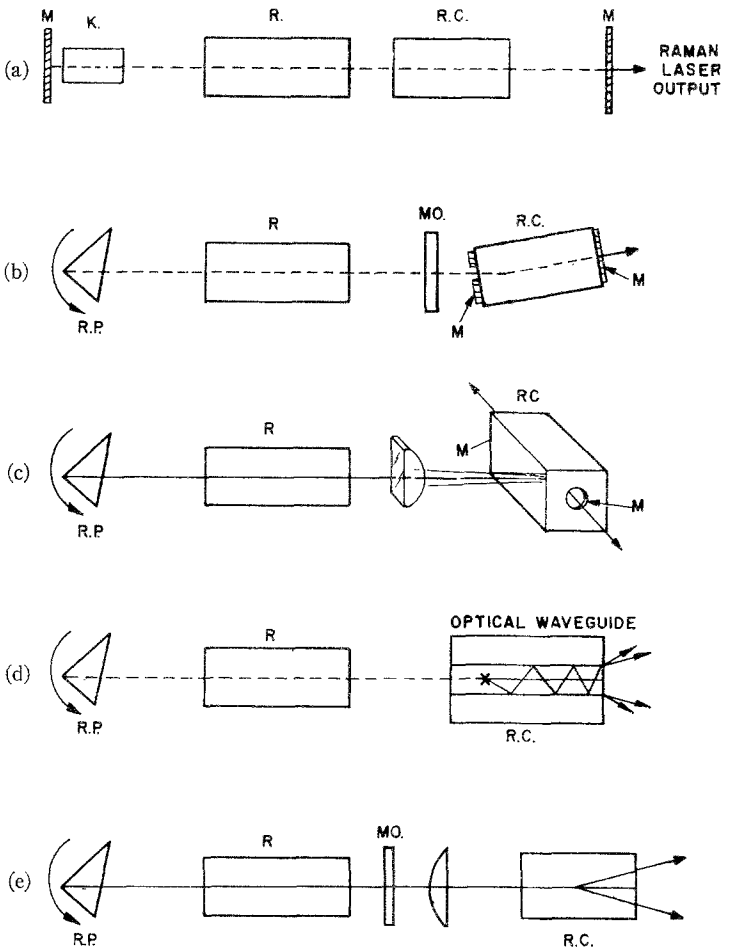

FIG. 3. Various experimental arrangements used for Raman laser: R, ruby; R. C., Raman cell; M, mirror; MO, mode selector; R. P., rotating prism; K, Kerr cell.

two beams decreases and consequently the Raman gain decreases. Therefore, the first Stokes radiation is obtained in a diffuse cone with an apex slightly larger than that of the exciting laser.

The anti-Stokes radiation requires the condition

$$\mathbf{k}_{AS1}=\mathbf{k}_L+\mathbf{k}_p \qquad (13)$$

to be satisfied. The three vectors can only be added in a particular fashion, as shown in Fig. 2(b). Thus the production of the anti-Stokes radiation is possible only in a certain direction making an angle to the exciting laser beam.

(4) The other striking feature is its efficiency.[8] Although the efficiency is dependent on exciting power, so far an efficiency of up to 60% in the first Stokes lines has been measured. This is an increase by a factor of about a million as compared to the spontaneous Raman scattering.

(5) As in ordinary lasers, there is also a line narrowing in Raman lasers, although it is not as spectacular as in the previous case. At higher powers, the linewidth is actually observed to increase. Sometimes the second- and third-order anti-Stokes lines can comprise[9] a very broad band up to 100 cm^{-1} wide, while the corresponding spontaneous Raman linewidth is 2 cm^{-1}. The increase in linewidth is at least partly due to the multimode effect, i.e., the interaction of various modes present in the exciting laser beam. This linewidth can, however, be reduced by using a proper mode selector to reduce the number of modes in the exciting laser. A Raman linewidth

[8] N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965).

[9] P. D. Maker and R. W. Terhune, Phys. Rev. **137**, A801 (1965).

of the order of 1 cm^{-1} has actually been achieved[10] by such a technique.

III. REVIEW OF LASER CAVITIES

The construction of Raman lasers using stimulated Raman scattering has been done using a variety of cavity arrangements:

(1) The first was that used by Woodbury and Ng.[11] In this arrangement, the Raman liquid is placed within the cavity of the exciting laser as shown in Fig. 3(a). The Raman laser gain in a cavity like this is small, but a considerable efficiency can be achieved by a large number of passes. The higher the reflection coefficient of the mirror, R, the larger is the number of passes. But the power emerging from the cavity decreases with increasing reflectivity of the mirror. The ratio of the power outside the cavity to that inside is given approximately by

$$P_0/P_i = 1-R. \tag{14}$$

For a mirror of reflectivity 99%, the output power is only 1% of the power inside the cavity.

(2) The off-axis Raman resonator, Fig. 3(b), used by Takuma and Jennings,[12] is interesting for the fact that, with selectively reflecting mirrors on the Raman cell, amplification may be achieved at a desired line. But the efficiency of Raman-laser action in this type of cavity is rather low, since the overlap between the exciting and Raman laser beams is small. The arrangement, Fig. 3(c), used by Dennis and Tannenwald[13] in which the oscillation is achieved at 90° to the pump radiation falls also in the above category.

(3) Recently, a novel cavity [Fig. 3(d)] has been used by Tang and Deutsch[14] in which the laser builds up in an optical waveguide. This arrangement shows the promise of high efficiency when using a long cell.

(4) By far, the most efficient conversion has so far been achieved in an arrangement [Fig. 3(e)] using[15] a lens to focus the exciting radiation on the Raman medium. The achievement of high efficiency in this arrangement can be immediately understood by recalling that the efficiency increases with the exciting power. Since the exciting power is increased many times at the focus of the lens, the efficiency achieved is very high. An efficiency of up to 60% within first Stokes line has been achieved by this configuration.

A simple analysis shows that the gain per pass through the focus is independent of the focal length of the lens. The theory of ideal focus for a focal length much larger than the aperture has been worked out in details in diffraction theory.[16] According to this theory, the region of high intensity near the focus can be described approximately by a plane wave traveling through a cylinder of length l and area a given by

$$l = (8f^2n/d^2)\lambda, \tag{15}$$

$$a = (\pi f^2/d^2)\lambda^2, \tag{16}$$

where f is the focal length and d is the beam diameter. The gain increases as the length [see Eq. (11)] and therefore as f^2. But the exciting intensity increases inversely as a, i.e., as f^2. Thus the gain by one pass through the focus is independent of the focal length used.

However, the choice of the focal length of the lens should be guided by other considerations. If too short a focal length is used, the exciting intensity may become so high that the power may be absorbed by multiphoton absorption and stimulated Brillouin scattering, etc. A focal length of 5 to 10 cm is found to be quite satisfactory. Since the value of l/a is independent of focal length and is approximately given[9] by

$$l/a = 3.5n/\lambda, \tag{17}$$

the gain [Eq. (11)] can be expressed in terms of single traversal through the focus

$$G = \exp[(2^6\pi^5 N\nu_R/3^2hc^2\Delta\nu_R)\alpha_{mn}{}^2 W_L \cdot 3.5n/\lambda], \tag{18}$$

where W_L is the total average power. The above gain equation agrees well with that derived by

[10] W. J. Jones and B. P. Stoicheff, Phys. Rev. Letters **11**, 657 (1964).
[11] E. J. Woodbury and W. K. Ng, Proc. IRE **50**, 2367 (1962).
[12] H. Takuma and D. A. Jennings, Appl. Phys. Letters **4**, 185 (1964).
[13] J. H. Dennis and P. E. Tannenwald, Appl. Phys. Letters **5**, 58 (1964).
[14] C. L. Tang and T. F. Deutsch, Phys. Rev. **138**, A1 (1965).
[15] R. W. Terhune, Bull. Am. Phys. Soc. **8**, 359 (1963).
[16] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Inc., New York, 1959), p. 434.

Maker and Terhune[9] (at least for gases where the refractive index is close to unity):

$$G=\exp[(14\pi NL\alpha^2 k_L k_R/chn_\omega{}^8\Delta\nu_R)W_L], \quad (19)$$

where L is the local-field correction factor which relates the applied macroscopic field to the field acting on the individual particle and k denotes the wave vector.

Maker and Terhune[9] assume that a gain of e^{30} (possibly more than e^{10} and less than e^{100}) determines the threshold of Raman-laser action. For strong Raman-active gases at about 50 atm pressure, Eq. (18) yields a gain in the above region with an exciting power of a few megawatts. This is in fair agreement with experimental observations.[17]

[17] R. W. Minck, R. W. Terhune, and W. G. Rado, Appl. Phys. Letters **3**, 181 (1963).

Kinematics—Quaternions—Spinors—and Pauli's Spin Matrices

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A simplified presentation of the relation between motion (space transformation) of a set of objects with spin and the induced (nonrelativistic) spinor transformation by development of quaternions from certain four-dimensional rotation matrices. These are interpreted as conformal transformations in three-space. Quaternions are essentially equivalent to Pauli spin matrices which can be generated by regarding a quaternion matrix as compound—i.e., a 2×2 matrix with 2×2 matrix elements. Explicit formulas with instructions for their use are given.

INTRODUCTION

THE apparent revival of interest in quaternions indicated by two recent publications[1,2] induces the present article on important fields of application of these hypercomplex number systems for physicists, and gives interpretations which do not seem to be widely known. The simple transition from quaternion units to Pauli spin matrices enhances their significance for modern physics.

The applications are to rotations in three- and four-dimensional spaces, to transformations in the Gauss plane of common complex numbers, and to (nonrelativistic) spinors. The interpretation is in the form of matrices of four rows and columns made up with only four independent parameters (instead of 16) and their products with column (or row) vectors of four components x_1, x_2, x_3, x_4, which are used to represent Cartesian coordinates in four-dimensional Euclidean space. The first three also may be used as coordinates in three-dimensional space when convenient.

In the interests of brevity, the following terminology is used: 2-plane means two-dimensional plane, which is necessarily situated in 3-space (i.e., three-dimensional space). Corresponding expressions apply to 3-plane, which is embedded in 4-space, etc. A 2-sphere is the surface of the sphere in 3-space. A 3-sphere in 4-space is a three-dimensional sphere embedded in four-dimensional space. The sphere $x_1^2+x_2^2+x_3^2+x_4^2=1$ is the unit 3-sphere in 4-space, etc.

I. ALGEBRAIC FORMALISM

The "common" quaternions of Hamilton are subject to the following multiplication table:

	i	j	k	1
i	-1	k	-j	i
j	-k	-1	i	j
k	j	-i	-1	k
1	i	j	k	1

(1)

The product of two of these "units" is obtained

[1] R. J. Stephenson, "Development of Vector Analysis from Quaternions," Am. J. Phys. **34**, 194 (1966).

[2] A. M. Bork, "Vectors Versus Quaternions—The Letters in *Nature*," Am. J. Phys. **34**, 202 (1966).