length is not well known, there is remarkable agreement between the calculations and the measured points.

The authors consider it likely that the use of Figs. 14 and 15 enable one to predict with reasonable assurance the pulsed and cw properties of air for frequencies from 100 Mc/sec to 100 kMc/sec over a range of pressure corresponding to an altitude variation of from 0 to 100 km, and over a very wide range of variation of container sizes.

PHYSICAL REVIEW VOLUME 130, NUMBER 5 1 JUNE 1963

Theory of Stimulated Raman Scattering

R. W. HELLWARTH
Hughes Research Laboratories, Malibu, California
(Received 22 October 1962)

Relations which describe the gain that is produced by the stimulated Raman scattering of intense light in a Raman-active material are developed entirely in terms of measurable material parameters (such as the ordinary Raman scattering cross sections). It is this gain which must overcome propagation losses in order to achieve the laser action that has been observed from this effect. Expressions for the gain are derived for scattering material either in thermal equilibrium or in certain nonequilibrium conditions. The dynamical equations governing the time behavior of the Raman-laser light are discussed.

WHEN various Raman-active liquids are placed inside an optical Fabry-Perot cavity and illuminated by light of frequency ω_α and intensity greater than a certain threshold, then coherent light builds up in the cavity at a frequency(s) ω_β which equals ω_α minus the frequency of a Raman-active vibration.[1,2] There is no (or negligible) resonant absorption at ω_α; all evidence indicates that stimulated Raman scattering is causing laser action.[2] Javan has proposed that a two-level Raman maser would be possible with no quantum states available other than an initial and final matter state[3]; he and Weber have calculated the rate at which such a two-level process would proceed.[3,4] However, the optical effect of interest here involves the normal Raman process with the mediation of large numbers of intermediate quantum states; the calculation of the matrix elements for this case would be extremely difficult. Therefore, we will content ourselves here with the development of a phenomenological theory to describe stimulated Raman scattering in terms of tabulated or measurable material parameters, namely ordinary Raman scattering cross sections. This treatment will be analogous to the treatment of ordinary lasers on the basis of absorption and fluorescence data.

The Raman process consists of (not necessarily in this order) annihilating a photon from a radiation mode α and creating a photon in another mode β (of different frequency), the energy difference being taken up by a transition in the scattering matter from a state i (of energy ϵ_i) to a state j (of energy ϵ_j) with $\epsilon_i+\hbar\omega_\alpha=\epsilon_j+\hbar\omega_\beta$. Unlike the case of fluorescence, there is no real intermediate state in this process which conserves energy. The possibility of laser action, or material gain, at ω_β arises from the values of the matrix elements for creating and annihilating photons. These matrix elements contribute a factor $n_\alpha(1+n_\beta)$ to the rate at which the process described above proceeds and a factor $n_\beta(1+n_\alpha)$ to the reverse process; n_α and n_β are the number of photons present in the incident and scattered modes, respectively. If we are considering the scattering of plane waves, then α represents the wave vector and polarization of the incident or pump radiation, β refers similarly to the scattered wave, which, by virtue of the term in the rate which is proportional to n_β, may become a growing wave in the pumped medium. When it is more convenient, α and β may refer to standing wave modes in a cavity; we will consider both cases. The scattering rate has, in addition to the factors mentioned above, a factor depending on matrix elements between various states in the medium $S_{ij}(\alpha,\beta)$ which also depends parametrically on ω_α and ω_β and on the spatial shapes of the modes α and β. Suppose the matter has probabilities P_i of being in the ith state. Then the average net rate of change of photon number in the scattered mode β arising from a given matter transition and incident mode α, is of the form

$$\dot{n}_\beta=n_\alpha(1+n_\beta)S_{ij}(\alpha,\beta)P_i-n_\beta(1+n_\alpha)S_{ji}(\beta,\alpha)P_j. \quad (1)$$

In (1) and what follows, the subscript j symbolizes those states for which $\epsilon_i+\hbar\omega_\alpha=\epsilon_j+\hbar\omega_\beta$. If the terms on the right-hand side of (1) which are proportional to n_β are positive, then they represent a contribution to

[1] E. J. Woodbury and W. K. Ng, Proc. I. R. E. **50**, 2367 (1962).
[2] Gisela Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, D. Weiner, and E. J. Woodbury, Phys. Rev. Letters **9**, 455 (1962).
[3] A. Javan, Bull. Am. Phys. Soc. **3**, 213 (1958); J. Phys. Radium **19**, 806 (1958).
[4] J. Weber, Rev. Mod. Phys. **31**, 681 (1959).

gain or laser action. A detailed theory shows that $S_{ij}(\alpha,\beta)$ equals $S_{ji}(\beta,\alpha)$; however, one can see that this must be true from (1) in order that thermal equilibrium can exist with $\dot{n}_\beta=0$. In general, there is a quasi-continuous set of Raman transitions $i\rightarrow j$ in the bulk matter which can contribute to scattering from α to β and, being concerned only with the total growth of n_β, we must sum over the transitions that contribute to it. Also, the incident light may come in many modes α, so that the total rate of scattering into mode β is of the form

$$\dot{n}_\beta=\sum_i\sum_\alpha[n_\alpha P_i-n_\beta P_j+n_\alpha n_\beta(P_i-P_j)]S_{ij}(\alpha,\beta). \quad (2)$$

The first term in brackets on the right-hand side of (2) describes what one would observe in a scattered wave β from an ordinary Raman scattering experiment with input intensities I_α (proportional to n_α) photons per second per unit area in the waves α. This first term may be easily converted to a relation for the differential scattering cross sections $\sigma_{\alpha\beta}$ [$\mathrm{cm^2(sr)^{-1}}$(unit-wavelength)$^{-1}$(volume of matter)$^{-1}$] for scattering from wave α into wave β:

$$\sigma_{\alpha\beta}=\sum_i P_i S_{ij}(\alpha,\beta)(n_\alpha/I_\alpha)\lambda_\beta^{-4}\ \mathrm{cm^{-2}}, \quad (3)$$

where λ_β is the wavelength (in the medium) of the scattered wave. n_α/I_α is a geometrical constant. [The functions in (3) may be compared with standard results for Raman cross sections[5] to obtain their exact dependence on the matter matrix elements, etc.] It is known that[5] (a) $\sigma_{\alpha\beta}$ varies slowly with angle in space (resembling typically a dipole radiation pattern for β modes) and (b) the $\sigma_{\alpha\beta}$ often exhibit peaks or resonances when $(\omega_\alpha-\omega_\beta)$ equals a Raman-active vibration frequency, but that they vary in a smooth manner with ω_α if $\omega_\alpha-\omega_\beta$ is fixed. [These $\sigma_{\alpha\beta}$ may depend on the temperature T of the material inasmuch as the relevant probabilities P_i vary with temperature in (3)]. The positions of these peaks are well tabulated for a large number of liquids, solids and gases; unfortunately for our purposes little attention has been paid to their absolute magnitudes.

The second term in brackets on the right-hand side of (2) is independent of the incident light intensity and represents an attenuation in the scattered waves due to Raman and Rayleigh scattering from them. This loss is generally small compared with other losses (impurity scattering, cavity wall losses, etc.) and in any case may be lumped together with them in a total loss term—$-w_\beta n_\beta$.

The last term in the brackets on the right-hand side of (2) is of crucial interest here as it describes a gain for mode β when $P_i>P_j$. We call the rate constant, which is the coefficient of n_β in this term, Γ_β. When α and β refer to traveling plane waves we can express Γ_β in terms of the cross sections $\sigma_{\alpha\beta}$ by using (3). In this case it is often more useful to consider the gain per cm $g_\beta=\Gamma_\beta/c_\beta$ (c_β is the velocity of light for the β wave), whence, (2) and (3) give

$$g_\beta=\sum_\alpha I_\alpha\sigma_{\alpha\beta}\lambda_\beta^4\{1-\exp[-\hbar(\omega_\alpha-\omega_\beta)/kT]\}/c_\beta\ \mathrm{cm^{-1}} \quad (4)$$

when the matter is in thermal equilibrium at temperature T. It is this gain which must overcome the average loss per cm to achieve laser action. In the experiments to date[1,2] the pump light I_α is on for such a short time ($<10^{-7}$ sec) and the exponential in (4) so small initially that one can conclude that the matter distribution function P_i did not vary significantly from that of thermal equilibrium during laser action; hence, Eq. (4) should describe g_β very well for these cases. Also, in these experiments the spectrum of the incident (well-collimated) light $I_\alpha(\omega_\alpha)$ was narrow compared to the resonances in $\sigma_{\alpha\beta}$; hence, no significant error results in approximating the sum on α in (4) by a single term as if all incident light were in a single mode. Were the incident light spectrum broad compared to the Raman line, only that portion of it roughly within a Raman linewidth would be effective in producing gain at a given frequency ω_β; but, in general, a coherent input is not necessary for coherent output. We have made guesses as to the magnitude of $\sigma_{\alpha\beta}$ from existing Raman data and find that (4) predicts gains consistent with observation[2]; however, further experiment is required for definitive numerical analyses of various materials.

In the case where one wishes to know Γ_β for stationary (standing wave) modes in terms of the cross sections $\sigma_{\alpha\beta}$, one may derive from (4) the imaginary part of the complex dielectric constant of the material and use it in Maxwell's equations with the mode boundary conditions to find Γ_β for the mode of interest. When the stationary mode can be thought of as consisting nearly of two oppositely traveling plane waves, then Γ_β (stationary mode) $\approx c_\beta g_\beta$ (running wave).

Let us consider the case where the distribution P_i is perturbed by laser action. Assume further that the Raman line in question is homogeneously broadened and can be thought of as arising from scattering from N nearly independent scattering particles per cc each having a (temperature-independent) cross section $s_{\alpha\beta}$ to scatter light from mode α to β by making a transition from a single initial state of energy E_1 to a single final state of energy E_2. (This would approximate the situation for many gases and some liquids.) Suppose N_1 and N_2 are the numbers of particles per unit volume in these states at any instant in time. Then the cross section $\sigma_{\alpha\beta}$ of (3) would equal $N_1 s_{\alpha\beta}$ and the gain per cm would be

$$g_\beta=\sum_\alpha I_\alpha s_{\alpha\beta}\lambda_\beta^4(N_1-N_2)/c_\beta\ \mathrm{cm^{-1}}. \quad (5)$$

[Note here that if the populations are inverted ($N_2>N_1$ for $E_2>E_1$), we may have gain at anti-Stokes lines ($\omega_\beta>\omega_\alpha$), which, according to (3), was not possible in thermal matter.]

[5] San-Ichiro Mizushima, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 26, p. 171.

In constructing dynamical equations to describe laser action it has been usually convenient to work with stationary modes. Following this here, we would interpret α and β accordingly and write from (2) equations for the β modes

$$\dot{n}_\beta=\Gamma_\beta[(N_1-N_2)n_\beta+N_1]-w_\beta n_\beta. \quad (6)$$

Coupling these with other differential equations describing how relaxation processes change N_2 and N_1 in the material would give a set of equations analogous to those used to describe stimulated fluorescence.[6,7] However, unlike for fluorescence, when the fraction of incident photons which is scattered becomes large (as has been observed to be possible[2]) the transparency of the material to the incident radiation falls drastically. The resulting reduction in n_α reacts back on n_β through Γ_β and extra equations (of a form depending on special circumstances) describing n_α must be coupled to those above. The relations (1)–(6) also apply when either the incident, or scattered, or both waves (α and β) are boson waves (such as sound) other than light.

The author wishes to thank Morrel Cohen for stimulating discussions and the Aspen Institute of Humanities, Physics Division, for their hospitality during part of this work.

[6] A. L. Schawlow and C. H. Townes, Phys. Rev. **112**, 1940 (1958).
[7] T. H. Maiman, Phys. Rev. **123**, 1145 (1961).

PHYSICAL REVIEW VOLUME 130, NUMBER 5 1 JUNE 1963

Charge Exchange and Dissociation of H^+, H_2^+, and H_3^+ Ions Incident on H_2 Gas*

G. W. McCLURE
Sandia Laboratory, Albuquerque, New Mexico
(Received 28 January 1963)

A unified set of measurements has been conducted of the yield cross sections for the various fast charged and neutral atomic particles produced in single collisions of H^+, H_2^+, and H_3^+ ions with H_2 gas molecules. The fast particles in question result from the dissociation or charge neutralization of the primary ions and have essentially the same velocity as the primary ions. The measurements cover the energy range from 3 to 100 keV for H^+ primaries, 3 to 120 keV for H_2^+ primaries, and 5 to 120 keV for H_3^+ primaries. The yield cross sections of the various dissociation fragments of H_2^+ and H_3^+ primaries are found to vary as much as 20% with changes in ion source operating conditions. It is proposed that this variation is due to changes in the population of internal energy states of the primary ions. Results obtained for H_2^+ primary ions are compared with the results of other investigators.

INTRODUCTION

THE destruction of fast H_2^+ ions (10 to 100 keV kinetic energy) in collisions with H_2 molecules has been shown to occur partly by electron capture with conversion of the primaries to H_2 neutrals or H+H pairs and partly by electronic excitation leading to conversion of the primaries to $H+H^+$ or H^++H^+ pairs.[1–6] The heavy products of the primary ion destruction move within a few degrees of the direction of the primary ions and have essentially the same velocity as the primary ions.[7] By the use of detectors such as the particle scintillation counter[4,5] and gas proportional counter,[6] these products are easily distinguishable from each other and are distinguishable from the relatively slow ionization and dissociation fragments of the target molecules. It is, therefore possible to determine the production cross section for each of the particle types and, with some manipulation of detector apertures,[4] to determine the reaction cross sections for the various destruction modes of H_2^+.

* This work performed under the auspices of the U. S. Atomic Energy Commission.
[1] K. K. Damodaran, Proc. Roy. Soc. (London) **239**, 382 (1957).
[2] C. F. Barnett, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Vol. 32, p. 398.
[3] N. V. Fedorenko, V. V. Afrosimov, R. N. Il'in, and D. M. Kaminker, Zh. Eksperim. i Teor. Fiz. **36**, 385 (1959) [translation: Soviet Phys.—JETP **9**, 267 (1959)].
[4] D. R. Sweetman, Proc. Roy. Soc. (London) **A256**, 416 (1960).
[5] J. Guidini, Compt. Rend. **253**, 829 (1961).
[6] Albert Schmid, Z. Physik **161**, 550 (1961).
[7] Dissociation fragments of primary ions of 5-keV kinetic energy are expected to appear at angles up to about 3° due to the mutual repulsion of the dissociating particles when electronic transitions occur to antibonding states. The maximum angular divergence associated with this process should vary as the inverse $\frac{1}{2}$ power of the primary ion energy. In some of the destructive collisions, a proton of the primary ion makes a nearly "head-on" impact with a proton of the target molecule. The first proton may then transfer a large fraction of its kinetic energy to the second and emerge from the collision at an angle much larger than that expected from the action of molecular antibonding forces. The cross section for these large-angle events can be roughly estimated from classical Rutherford scattering theory. Using the impulse approximation to the classical theory, the cross section for one of the protons of a 5-keV incident H_2^+ ion to scatter more than 3° on an H_2 molecule is $\simeq 1.6\times10^{-17}$ cm². This is negligibly small compared to either the H_2 production cross section ($\simeq 10^{-15}$ cm²) or the dissociation cross section ($\simeq 5\times10^{-16}$ cm²). Because of this circumstance, the "large-angle" collisions are usually ignored in accounting for the destruction of the primary ions.