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ON THE MECHANISM OF OPERATION OF A RAMAN LASER

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A classical analysis of forced combination (Raman) scattering is presented. A non-linear theory of a traveling wave amplifier based on this phenomenon is developed, and the condition for self-excitation of a Raman laser is derived.

THERE has been observed recently the phenomenon of self-excitation of light in a number of organic liquids placed under the influence of a pump — intense light waves of another frequency.[1,2] The frequency of the self-excited waves ω_s is connected with the frequency of the pump ω_p by the relation $\omega_p = \omega_s + \omega_0$, where ω_0 is one of the molecular vibration frequencies. An intimate connection between this phenomenon and the phenomenon of Raman scattering has been discovered, and it has been called forced Raman scattering. The generator itself has been called the Raman laser.

Hellwarth[3] has presented a simplified quantum-mechanical treatment of the phenomenon of forced Raman scattering based on the general structure of the relation between spontaneous and induced emission. A more complete theory based on a quantum kinetic equation was developed recently by Faĭn and Yashchin.[4]

In the quantum-mechanical description of the phenomenon of stimulated combination (Raman) scattering, molecular parameters (e.g., the effective cross section of a process) enter, which, as a rule, are determined either from experiment or by means of a classical analysis. This analysis in the theory of multi-atomic molecules is extremely fruitful and permits a large amount of data to be obtained about the structure of molecules.[5] Hence, in joining the classical theory of molecules with the theory of operation of the Raman laser a classical description of the phenomenon of stimulated Raman scattering would also be of interest.

This paper is devoted, first, to the classical description of the phenomenon of stimulated Raman scattering, and, second, to the theory of the Raman laser. It should be mentioned that since the structure of the classical and quantum expressions that are obtained for the polarization of a substance are the same (which indicates the validity of the classical approach), the theory of the laser is independent of the character of the description of the phenomenon of stimulated Raman scattering.

We shall consider the amplifying properties of a substance at a signal frequency ω_s, subjected to irradiation by intense light waves of another frequency ω_p being propagated in the same direction. If light waves are propagated through a substance with a nonlinear dependence of polarization on field, their behavior is described by the equation

$$\partial^2\mathbf{E}/\partial t^2 + 4\pi\partial^2\mathbf{P}/\partial t^2 + c^2 \operatorname{rot}\operatorname{rot}\mathbf{E} = 0, \qquad (1)^*$$

where **E** is the electric field and **P** is the polarization. Let these waves have the form

$$\mathbf{E} = \mathbf{E}_p e^{i(\omega_p t - \mathbf{k}_p\mathbf{r})} + \mathbf{E}_s e^{i(\omega_s t - \mathbf{k}_s\mathbf{r})} + \text{c.c.},$$

where $\mathbf{k}_s$ and $\mathbf{k}_p$ are wave vectors parallel to each other and the complex amplitudes $\mathbf{E}_p$ and $\mathbf{E}_s$ are in general slowly varying functions of the coordinates and time.

If the substance is homogeneous and fills the half-space $z > 0$, the amplitudes of the waves $\mathbf{E}_s$ and $\mathbf{E}_p$ in the established regime depend only on the coordinate z. In case of weak non-linearity of the polarization, the behavior of the amplitude is described by the equations

$$\cos\gamma \, d\mathbf{E}_m/dz = [i(k_m^2 - \omega_m^2/c^2)/2k_m]\mathbf{E}_m - 2\pi i k_m \mathbf{P}_m, \qquad (2)$$

where γ is the angle between the wave vector **k** and the z axis, and $\mathbf{P}_m$ is the amplitude of the wave of polarization of frequency ω_m:

$$\mathbf{P} = \mathbf{P}_p e^{i(\omega_p t - \mathbf{k}_p\mathbf{r})} + \mathbf{P}_s e^{i(\omega_s t - \mathbf{k}_s\mathbf{r})} + \text{c.c.}$$

Thus, in order to describe the behavior of the waves $\mathbf{E}_s$ and $\mathbf{E}_p$ it is necessary to determine the components of polarization $\mathbf{P}_s$ and $\mathbf{P}_p$.

Proceeding to this determination, we symbolize by x the normal coordinate of the vibrations of the atoms in a molecule of the substance and

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*rot = curl

by y the coordinate of electron vibrations. The frequency of the vibrations x correspond to the infrared region, and the electron vibration frequencies to the ultraviolet.

The phenomenon of Raman scattering is due to the presence of anharmonic terms in the expression U for the potential energy, which has the form

$$U = \tfrac{1}{2} Fx^2 + \tfrac{1}{2} fy^2 + \beta_1 x^3 + \beta_2 x^2 y + \beta x y^2 + \beta_3 y^3. \quad (3)$$

Here F and f are the "elasticities" of the bonds in the molecule; the coefficient β_1 determines the nonlinearity of the quasi-elastic vibrations x, β_2 the process of parametric excitation of the vibrations x by virtue of the excitation of the electronic motions, β_3 the nonlinearity of the electronic vibrations, which are responsible, in particular for the generation of optical harmonics, and finally, β determines the process of Raman scattering. We are interest only in this last process and shall retain only the anharmonic term proportional to β in Eq. (3).

In its purest form the Raman scattering phenomenon appears for fully symmetric vibrations x, which are absent in the absorption spectra. We limit ourselves to a consideration of only these vibrations. For simplicity, we also assume that the molecule is isotropic, so that the coordinate y is a vector.

Because of its symmetry, the vibration x is not coupled directly to the field of the light waves, and so the equation describing this vibration has the form

$$M\ddot{x} + H\dot{x} + Fx + \beta y^2 = 0. \quad (4)$$

Here M is the effective mass, H is a "friction" coefficient. If the frequencies ω_p and ω_s are lower by far than the frequencies of the electronic vibrations and correspond to the infrared region, then the equation of motion for the electronic vibration is "quasi-static" and has the form

$$f\mathbf{y} + 2\beta x \mathbf{y} = e\mathbf{E}. \quad (5)$$

Adding the relation $\mathbf{P} = eN\mathbf{y}$ to Eqs. (4) and (5), where e is the electronic charge and N is the number of molecules in unit volume, we obtain a closed system of equations for determining the amplitudes of $\mathbf{P}_s$ and $\mathbf{P}_n$.

Being interested only in stationary forced vibrations of the molecule, we shall seek solutions for the coordinates x and y in the form

$$x = Xe^{i(\omega_p - \omega_s)t} + \text{c.c.},$$

$$\mathbf{y} = \mathbf{Y}_p e^{i\omega_p t} + \mathbf{Y}_s e^{i\omega_s t} + \text{c.c.},$$

where it is not assumed in general that $\omega_p - \omega_s = \omega_0 = \sqrt{F/M}$. From Eqs. (4) and (5) follow the relations

$$f\mathbf{Y}_s + \beta X^+ \mathbf{Y}_p = e\mathbf{E}_s e^{-i\mathbf{k}_s \mathbf{r}}, \qquad f\mathbf{Y}_p + \beta X \mathbf{Y}_s = e\mathbf{E}_p e^{-i\mathbf{k}_p \mathbf{r}};$$

$$\omega_0 \delta (\Delta + i) X + (\beta/M) \mathbf{Y}_s^+ \mathbf{Y}_p = 0, \quad (6)$$

where

$$\Delta = [\omega_0^2 - (\omega_p - \omega_s)^2]/\omega_0 \delta, \qquad \delta = H/M.$$

From (6) are easily obtained relations for the amplitudes of $\mathbf{Y}_s$ and $\mathbf{Y}_p$, which when multiplied by eN become expressions for the amplitudes of the polarization $\mathbf{P}_s$ and $\mathbf{P}_p$:

$$2\pi \mathbf{P}_s = 2\pi \frac{e^2 N}{f} \mathbf{E}_s + \Gamma \frac{\mathbf{E}_s \mathbf{E}_p^+}{\Delta - i} \mathbf{E}_p,$$

$$2\pi \mathbf{P}_p = 2\pi \frac{e^2 N}{f} \mathbf{E}_p + \Gamma \frac{\mathbf{E}_p \mathbf{E}_s^+}{\Delta + i} \mathbf{E}_s, \quad (7)$$

where $\Gamma = 2\pi N \beta^2 e^4 / M f^4 \omega_0 \delta$. The lengths of the wave vectors $\mathbf{k}_s$ and $\mathbf{k}_p$ have not been specified yet. We choose these lengths such that the terms with a linear part of the polarization in Eq. (2) vanish.

Let us consider the interaction of the amplitudes of the waves $\mathbf{E}_s$ and $\mathbf{E}_p$ in the amplifier. The following equations can be obtained for these from Eqs. (2) and (7):

$$d\mathbf{E}_s/dz = \alpha_s (1 - i\Delta)(\mathbf{E}_p^+ \mathbf{E}_s)\mathbf{E}_p,$$

$$d\mathbf{E}_p/dz = -\alpha_p (1 + i\Delta)(\mathbf{E}_p \mathbf{E}_s^+)\mathbf{E}_s, \quad (8)$$

where $\alpha = k\Gamma / \cos\gamma (1 + \Delta^2)$. From Eqs. (8) it follows at once that if $\mathbf{E}_s$ and $\mathbf{E}_p$ are perpendicular to one another, there is no interaction of the waves.[1)] We introduce new coordinates z_1, z_2, z_3, choosing the z_3 axis along the direction of propagation of the waves. The vector differential equations (8) admit several first integrals of the form

$$E_{si} E_{sj}^+ / \alpha_s + E_{pi} E_{pj}^+ / \alpha_p = v_{ij} / \alpha_s, \qquad [\mathbf{E}_s \mathbf{E}_p] = \mathbf{B}. \quad (9)^*$$

Here E_{si} and E_{pi} are projections of $\mathbf{E}_s$ and $\mathbf{E}_p$ on the z_i axis, v_{ij} and $\mathbf{B}$ are scalar and vector constants.

The first of the relations in (9) can be interpreted as a law of conservation of the number of quanta. For example, in the case of linear polarization of the waves and i = j the first relation of (9) acquires the form

$$n_{si} + n_{pi} = n_{0i}, \quad (10)$$

where n_i is the number of light quanta of the corresponding frequency, polarized along the z_i axis,

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[1)] This is valid only for liquids consisting of isotropic molecules.

*$[\mathbf{E}_s \mathbf{E}_p] = \mathbf{E}_s \times \mathbf{E}_p$.

which pass through a unit area perpendicular to the z_3 axis.

Analysis of Eqs. (8) shows that if $\Delta \neq 0$ and $\mathbf{E}_s$ and $\mathbf{E}_p$ are not parallel to one another, the linearly polarized light becomes elliptically-polarized. In this the rotation of the vectors $\mathbf{E}_s$ and $\mathbf{E}_p$ proceeds to different sides. And when $\Delta = 0$ the linearly polarized light entering into the system remains the same, even if $\mathbf{E}_s$ and $\mathbf{E}_p$ are not parallel.

If the waves are linearly polarized, the value of the angle ψ between the vectors $\mathbf{E}_s$ and $\mathbf{E}_p$ is determined by the expression

$$\sin^2\psi = |\mathbf{B}|^2 / |\mathbf{E}_s|^2 \, |\mathbf{E}_p|^2.$$

The law for the change of the moduli of the amplitudes $\mathbf{E}_s$ and $\mathbf{E}_p$ can be obtained if use is made of the relation

$$|(\mathbf{E}_s\mathbf{E}_p^+)|^2 + |[\mathbf{E}_s\mathbf{E}_p]|^2 = |\mathbf{E}_s|^2 \, |\mathbf{E}_p|^2. \qquad (11)$$

We have

$$\frac{d}{dz}|\mathbf{E}_s|^2 = 2\alpha_c(|\mathbf{E}_p|^2 \, |[\mathbf{E}_s|^2 - |\mathbf{B}|^2),$$

$$\frac{d}{dz}|\mathbf{E}_p|^2 = -2\alpha_p(|\mathbf{E}_p|^2 \, |\mathbf{E}_s|^2 - |\mathbf{B}|^2). \qquad (12)$$

The solution of these equations has the form

$$|\mathbf{E}_s|^2 = v + \lambda \frac{1 - Ce^{-2\alpha_p\lambda z}}{1 + Ce^{-2\alpha_p\lambda z}},$$

$$|\mathbf{E}_p|^2 = \frac{\alpha_p}{\alpha_s}(v - |\mathbf{E}_s|^2). \qquad (13)$$

Here

$$\lambda = \sqrt{v^2 - \frac{\alpha_s}{\alpha_p}|\mathbf{B}|^2}, \qquad v = v_{11} + v_{22},$$

$$c = \frac{\lambda + (\alpha_s/\alpha_p)\,|\mathbf{E}_p(0)|^2}{\lambda - (\alpha_s/\alpha_p)\,|\mathbf{E}_p(0)|^2}.$$

The solutions (13), if considered for all values of z (not only positive ones), determine the transitions in energy from the state with $z = -\infty$, when $|\mathbf{E}_s|^2 = v - \lambda$, $|\mathbf{E}_p|^2 = (\alpha_p/\alpha_s)(v + \lambda)$ to the state at $z = +\infty$, when $|\mathbf{E}_s|^2 = v + \lambda$, $|\mathbf{E}_p|^2 = (\alpha_p/\alpha_s)(v - \lambda)$.

From this it is seen, first, that in the process of interaction of the waves energy passes from a wave at a higher frequency (the pump) to a wave with a lower frequency (signal). This explains why the amplifying properties appear only in a Stokes component of the Raman scattering and not in the anti-Stokes component. Second, in the general case not all the energy goes out from the pump wave to the signal wave. This is easy to understand, if one considers that the quanta of the signal field which have a polarization perpendicular to the polarization $\mathbf{E}_p$ do not take part in the interaction. It follows from the conservation law (10) that just as many quanta of the pump field will not take part in the interaction. Third, not all the energy going out from the pump wave passes into the signal wave since part of it is dissipated in exciting molecular vibrations. The maximum effectiveness of the energy transformation, corresponding to the case $B = 0$, equals $E_s^2(+\infty)/E_p^2(-\infty) = (\omega_s/\omega_p)$. It can be seen that it does not depend on the constants of the substance, on de-tuning of the frequencies, nor on the amplitude of the pump field.

It follows from Eq. (13) that the amplification of the signal depends on the distance passed through by the beam from the boundary, $l = z/\cos\gamma$, the polarizations of the waves, the energy of the pump at the amplifier input, the detuning Δ and the value of Γ, which depends on the constants of the substance.2) The bandwidth of the amplifier is determined by the quantity δ, i.e., the relaxation time of the vibration x.

As the amplitude of the signal wave grows, this wave itself begins to play the role of a pump and create amplifying properties at the frequency $\omega_{2s} = \omega_p - 2\omega_0$. As it grows, the self-excited wave at this frequency in its turn excites a wave of frequency $\omega_{3s} = \omega_p - 3\omega_0$, etc. In this way, as an intense light wave propagates through the dielectric a cascading decrease in its frequency takes place.

In order to go over to the Raman laser scheme, it is necessary to place the substance irradiated by the pump between two mirrors that are perpendicular to the beam of frequency ω_s. If the reflection coefficient of the mirrors is close to unity, the condition for self-excitation of the generator has the form

$$2\Gamma v > (1 - R^2)\,/\,kL, \qquad (14)$$

where L is the distance between the two mirrors. In this case the polarization of the oscillations of the generator will be the same as that of the pump.

In conclusion we remark that the Raman laser is a new kind of generator that can be related to but one of the types known in optics. Its energy of oscillation is proportional to $(\omega_s/\omega_p)E_p^2$, which relates such generators to parametric ones.[7] At the same time they have also a number of

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2) The coefficient Γ is equal in order of magnitude to 10^{-9} cgs esu, which agrees with the experimental data.[2]

essential distinctions from parametric generators —the fulfillment of a rather strict dispersion relation is not required, the self-excitation coefficient is determined by the square and not the first power of the amplitude of the pump wave, and others.

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