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INDUCED RAMAN SCATTERING IN STRONG FIELDS

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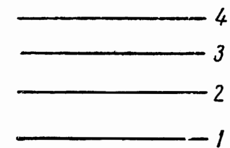
The problem of the splitting of the Stokes lines in induced Raman scattering is considered. The magnitude of the field strengths of the exciting wave is considered. It is shown that there is nonsymmetric broadening of the Stokes line in strong fields.

1. At the present time, there is a sufficiently complete quantum theory of induced (Raman) scattering in fields close to the threshold.[1,2] In the works of Javan[1] and Khronopulo,[3] the effects of saturation in strong fields are also considered.

For high powers of the exciting wave, several additional effects are possible, which are associated with the nonequivalence of the vibration levels of the molecule (see the figure). If the differences $\omega_{21} - \omega_{32}$, $\omega_{32} - \omega_{43}, \ldots$ are large, then it is not necessary to take into account the transitions 2 – 3 and 3 — 4 under the action of not very strong fields with frequencies ω and $\omega - \omega_{21}$. This leads to complete "saturation," that is, to a limitation of the growth rate of the Stokes component of the light. Such an approach is not correct in those cases in which these differences are comparable with the half-width of the Stokes lines; then, in strong fields, the transitions 2 – 3 and 3 – 4 play a significant role, while the kinetic equation for the density matrix ceases to be valid in that form in which it is used in the cited papers. In those cases in which these differences significantly exceed the half-width of the level, it is interesting to study the problem of the excitation of the Stokes frequency $\omega - \omega_{32}$, that is, the problem of the conversion of the Stokes line into a doublet. This frequency can be excited in strong fields in the case of an appreciable population in the second level, maintained by induced (and thermal) transitions 1 – 2.

The classical theory of induced combination scattering (ICS)[4,5] is in practice equivalent to consideration of an almost equidistant set of vibration levels. The nonlinearity of the oscillator in such a theory can reduce to the slowing (but not to the limiting) of the growth rate of the Stokes component of the light, and the appearance of a Stokes doublet is not possible in this case. In the present research, a quantum mechanical consideration of ICS is given for large pump powers with account of the vibration transitions 2 – 3, 3 — 4, and so forth, when the differences $\omega_{21} - \omega_{32}$, $\omega_{32} - \omega_{43}$ are appreciably greater than the half-width of the level. Together with "saturation" and the transition of the Stokes line into a doublet at high pump powers, a shift in the Stokes frequency is also possible.



System of vibrational levels of a molecule.

2. We shall begin with the kinetic equation for the density matrix in the energy representation:

$$\frac{\partial \rho_{kl}}{\partial t} + \left(\frac{1}{\tau_{kl}} + i\omega_{kl}\right)\rho_{kl} = -\frac{i}{\hbar}\sum_j (V_{kj}\rho_{jl} - \rho_{kj}V_{jl}),\ k \neq l;$$

$$\frac{\partial \rho_{ll}}{\partial t} = -\frac{i}{\hbar}\sum_j (V_{lj}\rho_{jl} - \rho_{lj}V_{jl}) + \sum_j (W_{jl}\rho_{jj} - W_{lj}\rho_{ll}), \quad (1)$$

where $V_{ij} = -\mu_{ij}E$ and for simplicity, we set $\mu_{12} = 0$. If a Stokes wave is excited in the medium irradiated by optical pumping, the field can be represented in the form

$$E = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + \text{c.c.},$$

where $\omega_1 + \omega_2 = \omega_{21} + \Delta_{12}$; Δ_{12} is small ($-\omega_2$ is the Stokes frequency). Let E_1 and E_2 be not too large, such that the transitions 2 – 3 are of low probability. Then it is important to retain in the system (1) only equations containing the indices 1 or 2. Inasmuch as the relaxation time τ_{ij} is at least three orders of magnitude smaller than the time of the laser pulse, one can look for a solution for the nondiagonal elements in the steady state:

$$\rho_{1l} = R_{1l}e^{i\omega_1 t} + r_{1l}e^{i\omega_2 t} + R_{1l}^- e^{-i\omega_1 t} + r_{1l}^- e^{-i\omega_2 t},$$

$$\rho_{l2} = R_{l2}e^{i\omega_1 t} + r_{l2}e^{i\omega_2 t} + R_{l2}^- e^{-i\omega_1 t} + r_{l2}^- e^{-i\omega_2 t}, \quad (2)$$

$$\rho_{12} = \rho_{12}^+ e^{i(\omega_1+\omega_2)t} + \rho_{12}^- e^{-i(\omega_1+\omega_2)t}.$$

Here it is not necessary to consider that ρ_{ll} and E_n are constant. It is assumed only that they change slowly, so that in practice ρ_{1l} and ρ_{2l} follow their changes almost instantaneously.

In the expression for ρ_{12}, one can neglect the term ρ_{12}^- in comparison with the resonance ρ_{12}^+; this leads to the result that the quantities R^- and r^- contain only a linear part. For R, r, and ρ_{12}^+, we get [after substitution of (2) in (1)] the following relations:

$$R_{1l}(\omega_1+\omega_{1l}-i/\tau_{1l}) = \hbar^{-1}\mu_{1l}E_1(\rho_{ll}-\rho_{11}) - \hbar^{-1}E_2^*\mu_{2l}\rho_{12}^+,$$
$$r_{1l}(\omega_2+\omega_{1l}-i/\tau_{1l}) = \hbar^{-1}\mu_{1l}E_2(\rho_{ll}-\rho_{11}) - \hbar^{-1}E_1^*\mu_{2l}\rho_{12}^+,$$
$$R_{l2}(\omega_1+\omega_{l2}-i/\tau_{l2}) = \hbar^{-1}\mu_{l2}E_1(\rho_{22}-\rho_{ll}) + \hbar^{-1}E_2^*\mu_{l1}\rho_{12}^+,$$
$$r_{l2}(\omega_1+\omega_{l2}-i/\tau_{l2}) = \hbar^{-1}\mu_{l2}E_2(\rho_{22}-\rho_{ll}) + \hbar^{-1}E_1^*\mu_{l1}\rho_{12}^+,$$
$$\rho_{12}^+(\Delta-i\delta) = \hbar^{-1}\sum_l[\mu_{1l}(E_1r_{l2}+E_2R_{l2}) - \mu_{l2}(E_1r_{1l}+E_2R_{1l})]. \qquad (3)$$

In the determinant of the set (3), only the diagonal elements and the last row and column differ from zero. Solution of the set (3) for ρ_{12}^+ has the form

$$\rho_{12}^+ = \frac{\operatorname{Det}\rho}{\operatorname{Det}}; \quad \operatorname{Det}\rho = -\sum_l\sum_{s=1}^4 a_{n,sl}\,b_{sl};$$
$$\operatorname{Det} = -\sum_l\sum_{s=1}^4 a_{n,sl}a_{sl,n} + a_{nn}, \qquad (4)$$

where $a_{n,sl}$ are coefficients in Eqs. (3), while the column of three terms is denoted by b. In what follows, we shall neglect the quantities i/τ and Δ in the nonresonant terms. Then, by making use of the relation

$$\frac{1}{\omega_1+\omega_{1l}-i/\tau_{1l}} + \frac{1}{\omega_2+\omega_{1l}-i/\tau_{1l}} \approx -\frac{1}{\omega_1+\omega_{l2}-i/\tau_{l2}} - \frac{1}{\omega_2+\omega_{l2}-i/\tau_{l2}} \approx -\frac{\omega_{l1}+\omega_{l2}}{(\omega_{l2}+\omega_1)(\omega_{l2}+\omega_2)}, \qquad (5)$$

we obtain from (3) and (4)

$$\operatorname{Det}\rho = -\frac{E_1E_2}{\hbar^2}(\rho_{11}-\rho_{22})\sum_l\frac{\mu_{1l}\mu_{l2}(\omega_{l2}+\omega_{l1})}{(\omega_{l2}+\omega_1)(\omega_{l2}+\omega_2)},$$

$$\operatorname{Det} = \Lambda_{12} - i\delta,$$

$$\delta = \frac{1}{\tau_{21}}; \quad \Lambda_{12} = \Delta_{12} - \frac{1}{\hbar^2}\sum_l\left(\frac{|\mu_{l2}E_2|^2}{\omega_1+\omega_{1l}} + \frac{|\mu_{l2}E_1|^2}{\omega_2+\omega_{1l}} + \frac{|\mu_{l1}E_2|^2}{\omega_1+\omega_{l2}} + \frac{|\mu_{l1}E_1|^2}{\omega_2+\omega_{l2}}\right). \qquad (6)$$

For the diagonal elements, by substituting (2) in (1), we get the relation

$$\frac{\partial\rho_{11}}{\partial t} = -\frac{i}{\hbar}\sum_l[\mu_{l1}(E_1R_{1l}+E_2r_{1l}) + \text{c.c.}] + \sum_l(W_{l1}\rho_{ll} - W_{1l}\rho_{11}),$$
$$\frac{\partial\rho_{22}}{\partial t} = -\frac{i}{\hbar}\sum_l[\mu_{2l}(E_1^*R_{l2} + E_2^*r_{l2}) + \text{c.c.}] + \sum_l(W_{l2}\rho_{ll} - W_{2l}\rho_{22}) \qquad (7)$$

and so forth. Taking (3) and (5) into account, we find

$$\frac{\partial\rho_{11}}{\partial t} = -\frac{iE_1^*E_2^*}{\hbar^2}\sum_k\frac{\mu_{k1}\mu_{k2}(\omega_{k1}+\omega_{k2})}{(\omega_{k2}+\omega_1)(\omega_{k2}+\omega_2)}\rho_{12}^+ + \text{c.c.} + \sum_l(W_{l1}\rho_{ll} - W_{1l}\rho_{11}). \qquad (8)$$

Substituting ρ_{12}^+ from (4) and (6) in (8), we get

$$\frac{\partial\rho_{11}}{\partial t} = -\frac{2\delta\sigma_{12}}{\hbar^4(\delta^2+\Lambda_{12}^2)}(\rho_{11}-\rho_{22})|E_1|^2|E_2|^2 + \sum_l(W_{l1}\rho_{ll} - W_{1l}\rho_{11}),$$
$$\sigma_{12} = \left|\sum_l\frac{\mu_{1l}\mu_{l2}(\omega_{l1}+\omega_{l2})}{(\omega_{l2}+\omega_1)(\omega_{l2}+\omega_2)}\right|^2. \qquad (9)$$

Similarly,

$$\frac{\partial\rho_{22}}{\partial t} = \frac{2\delta\sigma_{12}}{\hbar^4(\delta^2+\Lambda_{12}^2)}(\rho_{11}-\rho_{22})|E_1|^2|E_2|^2 + \sum_l(W_{l2}\rho_{ll} - W_{2l}\rho_{22}). \qquad (10)$$

In the expressions for $\partial\rho_{ll}/\partial t$, the term which depends on the field falls out for $l \neq 1, 2$; therefore, one can assume that the ρ_{ll} are small for $l \neq 1, 2$, as also in the absence of the field. In the general case, the amplitudes E_1 and E_2 depend on ρ_{ll} and it is necessary to solve Eqs. (9) and (10) together with the Maxwell field equations. However, Eqs. (9) and (10) can easily be solved if the field is assumed to be given. This is permissible if $1/W_{21}$ is significantly smaller than the period of the laser pulse, that is, when one can seek a steady state for ρ_{11} and ρ_{22}. Denoting

$$B_{12} = \frac{2\delta\sigma_{12}}{\hbar^4(\Lambda_{12}^2+\delta^2)}|E_1|^2|E_2|^2$$

and for simplicity neglecting ρ_{ll} when $l \neq 1, 2$, we find from the condition $\partial\rho_{11}/\partial t \approx \partial\rho_{22}/\partial t \approx 0$ that

$$\rho_{11} = \frac{B_{12}+W_{21}}{2B_{12}+W_{21}+W_{12}}; \quad \rho_{22} = \frac{B_{12}+W_{12}}{2B_{12}+W_{21}+W_{12}}. \qquad (11)$$

The quantity B_{12} characterizes the probability of the induced transition 1 – 2.

We now find the nonlinear part of the polarization $P'(\omega_2)$ per molecule. For this purpose, we make use of the expression $P = \mathrm{Sp}\, \rho\mu$ and Eqs. (2), (3), and (5). We have

$$P'(\omega_2) = \sum_k \frac{\mu_{2k}\mu_{k1}(\omega_{k1}+\omega_{k2})}{(\omega_{k2}+\omega_1)(\omega_{k2}+\omega_2)} \rho_{12}^+.$$

Substituting the expression for ρ_{12}^+ from (4) and (6) in this, we obtain

$$P'(\omega_2) = -\frac{\Lambda_{12}+i\delta}{\Lambda_{12}^2+\delta^2} \frac{(\rho_{11}-\rho_{22})}{\hbar^3} \sigma_{12}|E_1|^2 E_2. \quad (12)$$

If $E_2 = \mathcal{E}_2 \exp(i\omega_2 z/c)$, where $\mathcal{E}_2$ depends weakly on the coordinate z, then one can get for $\partial\mathcal{E}_2/\partial z$ from Maxwell's equations (see, for example, [6])

$$\frac{\partial \mathcal{E}_2}{\partial z} \approx -\alpha_2 \mathcal{E}_2 - i2\pi \frac{\omega_2}{c} P'(\omega_2),$$

where α_2 is the linear absorption at the frequency ω_2. Then

$$\frac{\partial \mathcal{E}_2}{\partial z} \approx \left(-\alpha_2 - 2\pi \frac{\omega_2}{c} \frac{\delta(\rho_{11}-\rho_{22})}{\hbar^3(\Lambda_{12}^2+\delta^2)} \sigma_{12}|E_1|^2\right) \mathcal{E}_2. \quad (12')$$

This equation determines the behavior of the wave component $\mathcal{E}_2$ in the medium under consideration.

3. If a similar analysis is made of the transitions 2 – 3, one must add equations for ρ_{23}, ρ_{2l}, and ρ_{l3} to the set (2). These equations are not "coupled" with Eqs. (2), so that the solutions for ρ_{12}, ρ_{1l}, ρ_{l2} are not changed, while the solutions for ρ_{23}, ρ_{2l}, and ρ_{l3}, which are written in the form (3), have exactly the same form with replacement of the indices 1, 2 by 2, 3. Equation (9) is also unchanged with respect to transitions 2 – 3, while in Eq. (10), a term $-B_{23}(\rho_{22}-\rho_{33})$ is added for $\partial\rho_{22}/\partial t$, which takes into account these transitions (B_{23} is obtained from B_{12} by replacing the indices 1, 2 by 1, 3). The term of opposite sign is added to $\partial\rho_{33}/\partial t$. In the expression for $P'(\omega_2)$ there appears an additional component from (12) by the substitution 1, 2 → 2, 3. This process can be continued for the transitions 3 – 4 and so forth.

However it is necessary to take into account such additional terms only when saturation effects are considered. It must be expected that the splitting of the Stokes line takes place before the saturation effects play an important role, that is, before ρ_{22} becomes large, close to $1/2$. For consideration of the splitting effect, we represent the field in the form of three waves

$$E = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + E_3 e^{i\omega_3 t} + \text{c.c.},$$

where $-\omega_3 = \omega_1 - \omega_{32} - \Delta'_{23}$, and we add an equation for ρ_{23}, ρ_{2l}, and ρ_{l2} to the system (2). Here, one must take into account in ρ_{2l} and ρ_{l3} only terms with frequencies ω_1 and ω_3, and in ρ_{23} the term $\rho_{23}^+ e^{i(\omega_1+\omega_3)t}$. These equations are again not coupled with Eqs. (2). For $\partial\mathcal{E}_3/\partial z$ in such a form, as above, we get the expression

$$\frac{\partial \mathcal{E}_3}{\partial z} = \left(-\alpha_3 - 2\pi \frac{\omega_3}{c} \frac{\delta(\rho_{22}-\rho_{33})}{\hbar^3(\Lambda_{23}^2+\delta^2)} \sigma_{23}|E_1|^2\right) \mathcal{E}_3. \quad (12'')$$

For powers of the order of threshold, one can assume in (12′) $\Lambda_{12} = 0$ and $\rho_{11} - \rho_{22} \approx 1$. Similarly, for powers $|E_1|^2$ and $|E_2|^2$, which are the thresholds for excitation of the second Stokes component, $\Lambda_{23} = 0$ and $\rho_{23} - \rho_{33} \approx \rho_{22}$.

By comparing Eqs. (12) and (12′), and assuming $\sigma_{12} \approx \sigma_{23}$, we get for the power $|E_1|_d^2$ which is threshold for excitation of the doublet,

$$\rho_{22}|E_1|_d^2 \approx |E_1|_t^2,$$

where $|E_1|_t^2$ is the threshold for excitation of the primary Stokes component. Substituting ρ_{22} from (11), we have

$$\frac{B_{12}+W_{12}}{2B_{12}+W_{21}+W_{12}} |E_1|_d^2 \approx |E_1|_t^2. \quad (13)$$

The ratio W_{12}/W_{21} for $\omega_{21} \approx 1000\ \text{cm}^{-1}$ is smaller than 0.01. Therefore, one can neglect the quantity W_{12} in (13); for very low frequencies, the quantity W_{12} can exceed B_{12}; then the condition (13) takes the simple form:

$$\frac{W_{12}}{W_{21}}|E_1|_d^2 = \exp\left(-\frac{\hbar\omega_{21}}{kT}\right)|E_1|_d^2 \approx |E_1|_t^2.$$

As has already been pointed out, the splitting of the Stokes component must continue up to the case in which ρ_{22} becomes close to $1/2$, that is, when $2B_{12}$ is still small in comparison with W_{21}; consequently, one can neglect the term $2B_{12}$ in the denominator of (13).

It will be shown below that, at least in some cases, one can assume that $\Lambda_{12}^2 < \delta^2$. After all the simplifications, we get in place of (13):

$$\frac{2\sigma_{12}}{\delta\hbar^4 W_{21}} |E_1|_d^4 |E_2|_d^2 \approx |E_1|_t^2. \quad (13')$$

The quantity $\sigma_{12}/\delta\hbar^4$ was estimated on the basis of data obtained experimentally by McClung and Weiner [7] and the well known expression for the intensity of spontaneous combination scattering. For substances studied in [7], $\sigma_{12}/\delta\hbar^4 \approx 10^{-6}$ cgs esu. The quantity $|E_2|^2$ can amount to several times ten percent of $|E_1|^2$. We take $|E_2|^2 = 0.1 \times |E_1|^2$. Then, if we set $W_{21} = 10^{10}\ \text{sec}^{-1}$, $\sigma_{12}/\delta\hbar^4 = 10^{-6}$ (for example, for benzene):

$$|E_1|_d^6 \approx 5 \cdot 10^{-16} |E_1|_t^2.$$

Inasmuch as $|E_1|_d^2 > |E_1|_t^2$, the threshlod for the excitation of the Stokes doublet, the value of E_1 does not exceed r × 10^6 V/cm in the case of benzene. Such a field is quite realizable experimentally.

Strictly speaking, the given estimates are valid only for ICS in a resonator. For a focused beam, the field E_1 in some region can exceed 5×10^6 V/cm, but either E_2 is shown to be too small (inasmuch as the light of the Stokes frequency is not focused) or the volume in which the condition (13) is satisfied will be small, so that the wave E_3 is not excited. Moreover, only for a resonator can one show rigorously that $\Lambda_{12}^2 < \delta^2$; this must be kept in mind in the realization of the splitting of the Stokes components in ICS.

4. Let us now consider the effect of the pump power on the ICS spectrum. For this case, it is necessary to estimate the value of Λ_{12}^2 in the denominator of (12). We set $\mu_{1l} \approx \mu_{2l}$ and use the equality [6]

$$|E_1|^2 = A + \omega_1 |E_2|^2 / \omega_2 \approx A - |E_2|^2.$$

Then Eq. (6) for Λ_{12} is easily transformed into

$$\Lambda_{12} = \Delta_{12}' + \frac{2|E_2|^2}{\hbar^2} \sum_l \frac{\mu_{1l}\mu_{l2}(\omega_1 - \omega_2)}{(\omega_{l2} + \omega_1)(\omega_{l2} + \omega_2)},$$

$$\Delta_{12}' = \Delta_{12} - \frac{A}{\hbar^2} \sum_l \frac{\mu_{1l}\mu_{l2}(\omega_1 - \omega_2)}{(\omega_{l2} + \omega_1)(\omega_{l2} + \omega_2)}. \quad (14)$$

A wave is excited such that in the main part of the resonator the quantity $|\Lambda_{12}|$ is minimal; in this part of the resonator, in any case,

$$\Lambda^2 \leqslant 4 \frac{|E_2|^4}{\hbar^2} \left| \sum_l \frac{\mu_{1l}\mu_{l2}(\omega_1 - \omega_2)}{(\omega_{l2} + \omega_1)(\omega_{l2} + \omega_2)} \right|^2. \quad (14')$$

If μ_{1l} take the largest value for the electronic transitions, then $\omega_{l1} + \omega_{l2} \gtrsim \omega_1 - \omega_2$ and the square of the sum in (14′) does not exceed the value σ. We then have

$$\delta \geqslant W_{21} > 2B_{12} = \frac{4\delta}{\delta^2 + \Lambda_{12}^2} \frac{\sigma_{12}}{\hbar^4} |E_1|^2 |E_2|^2 \geqslant \frac{\delta}{\delta^2 + \Lambda_{12}^2} \Lambda_{12}^2.$$

One can assume that $\delta > 2\delta\Lambda_{12}^2/(\delta^2 + \Lambda_{12}^2)$, that is, $\delta^2 > \Lambda_{12}^2$. If the beam is focused, the quantity Λ_{12} changes along its axis. For simplicity, neglecting $|E_2|^2$ in comparison with $|E_1|^2$, and assuming $\mu_{1l} \approx \mu_{2l}$, we get for Λ_{12} from (6):

$$\Lambda_{12} = \Delta - \frac{|E_1|^2}{\hbar^2} \sum_l \frac{(\omega_1 - \omega_2)\mu_{1l}\mu_{l2}}{(\omega_{l2} + \omega_1)(\omega_{l2} + \omega_2)}. \quad (15)$$

It is natural to assume that the pump field excites a wave of frequency $-\omega_2$ such that the quantity Λ_{12}^2 is small in that part of the space where E_1 is maximum; that is, where the amplification is greatest. Here too, there are regions in which the inequality $\delta^2 > \Lambda_{12}^2$ is satisfied. The condition of minimum value of Λ_{12}^2 determines the magnitude and sign of the detuning Δ_{12}.

The sign of the sum on the right hand side of (15) is determined by which of the components—with $\omega_1 > -\omega_2$ or with $\omega_{l2} < -\omega_2$—gives the principal contribution. In order of magnitude, this sum is close to $\sigma^{1/2}$. We assume for the time being that this sum is positive and consider the change in Δ_{12} with time. The pump light pulse has a bell shape. In the period of growth of $|E_1|^2$ the detuning Δ_{12} increases, that is, the frequency decreases. A large part of the energy of the Stokes wave is radiated when $|E_1|^2$ is maximal and approximately constant. In this time, the frequency $-\omega_2$ is minimal. In the passage of the rear front of the pulse the frequency $-\omega_2$ again increases. The frequency $-\omega_2$ manages to follow the change in $|E_1|^2$ inasmuch as the duration of the fronts amounts to 10^{-9} sec, while the rise time of E_2 is 10^{-11} sec. On passage of the fronts of the pulse the lesser part of the energy is radiated. Therefore the ICS spectrum is sharply limited on the red side and becomes smeared in the blue. If the sum in (15) is negative, smearing of the spectrum on the red side should be noted. Such a smearing of the spectrum also takes place in the resonator. In this case, there is the possibility of determining the sign of the sum in (15) and of drawing a conclusion as to which levels play the dominant role in combination light scattering.

At the focus of the beam, the effect is complicated by the strong spatial inhomogeneity of the field. Let us consider a space in which the field is above the threshold. Most of the space is taken up by a region in which the field is too small to give an appreciable shift in the frequency $-\omega_2$. In this region, waves are excited whose frequency $-\omega_2$ is maximal (if the sum in (15) is positive). As the exciting field grows the Stokes component has an ever smaller frequency. This leads to a smearing of the spectrum on the red side. Therefore, at the focus of the pumping beam the spectrum should be smeared on both sides; however, this smearing is not symmetric.

Let us estimate the value of the field E_1 for which $\Delta_{12}^2 \approx \delta^2$. Taking the sum in (14) equal to $\sigma^{1/2}$, we have

$$\Delta_{12}^2 / \delta \approx \delta = \sigma_{12} |E_1|^4 / \delta\hbar^4 \approx 10^{-6} |E_1|^4.$$

If $\delta = 10^{11}$ sec^{-1}, then $E_1 \approx 6 \times 10^6$ V/cm. In view of the strong dependence on $|E_1|^2$, the smearing of the ICS lines in strong fields can be considerable.

We note that upon excitation of ICS in a resonator, the principal role in the phenomenon of saturation is played by the decrease in the difference $\rho_{11} - \rho_{22}$ (inasmuch as $\Lambda_{12}^2 < \delta^2$). At the focus of the beam, depending on the value of the sum in (15), the principal role of saturation can be played by the term Λ_{12}^2 in the denominator of (12).

5. It is thus shown that in ICS the appearance of the doublet Stokes lines is possible at fields determined by the condition (13). This field is less than that for which the effects of saturation are significant. For large but achievable pump power, the Stokes line should be diffuse. The character of the diffusion allows us to estimate what energy levels made the greatest contribution to the combination light scattering. For excitation of ICS in a resonator the chief role in the saturation effect is played by the decrease in the population of the first level and the increase in the population of the second. At the focus of the beam, the decrease of $\rho_{11} - \rho_{12}$ can also play an important role at least in the region of maximal field. In this case, calculation of the transitions $2 - 3$ is essential, if the anharmonicity is comparable with the width of the Stokes line.

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