Effect of four-wave parametric processes on the dynamics of the Stokes components of stimulated Raman scattering

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Generation of SRS Stokes components under biharmonic pumping is investigated theoretically and experimentally. It is shown that for experimental observation of the weak-pump Stokes component ω_1 the threshold ω_2 of the strong-pump SRS must be exceeded. In this case it is possible to observe the Stokes component ω_{1S} of radiation whose intensity is lower than the SRS threshold by several orders of magnitude. This is due to a four-wave parametric process of the type $\omega_1-\omega_{1S}=\omega_2-\omega_{2S}$. Efficient transformation of the weak pump is observed at lengths that exceed considerably (by 10^2 times) the linear synchronism length $(\delta k)^{-1}=[(k_2-k_{2S})-(k_1-k_{1S})]^{-1}$. It is shown that the reason why the process is not critical with respect to wave detuning is the phase locking of the interacting waves. The spatial scale of the weak transformation is determined by the length of combination transformation of strong pumping. The effect of the relation between the frequencies and intensity of the biharmonic pumping components and the width of the weak-pump spectrum on the efficiency and dynamics of the process is investigated. The broadband emission of a low-power dye laser that can be tuned in a band of ~25 nm was transformed in the experiment into long-wave radiation. It is shown that the weak-pump Stokes component may be obtained by two-photon absorption of the strong-pump radiation. Owing to the high efficiency and weak dependence on the wave detuning, the processes under consideration can be used to generate tunable infared radiation.

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I. INTRODUCTION

We have already reported[1] an investigation of the influence of parametric processes on the generation of Stokes components of stimulated Raman scattering (SRS) under biharmonic pumping. This process was investigated earlier for purposes of nonlinear spectroscopy[2,3] when Raman interaction of the fields is insignificant and the linear-synchronism conditions are satisfied (by choice of the appropriate geometry of the interacting beams). In our experiments, generation was observed in collinear beams at an appreciable value of the wave detuning. It was shown that four-wave parametric interaction (FPI) can nevertheless make the main contribution to the generation of the Stokes component by a weaker pump component. This is due to the locking of the phase of the generated Stokes wave. The characteristic length of the parametric interaction of the wave is determined not by the wave detuning, but by the scale of conversion of the strong component (i.e., exceeding the SRS threshold) of the pump into its Stokes component. As a result, the weak-pump power needed to produce its Stokes component can be lowered by several orders of magnitude.[1]

The foregoing properties of the discussed process make it promising for an effective conversion of the frequency of low-power frequency-tunable lasers in the infrared band. The low sensitivity to the magnitude of the wave synchronism can permit a large frequency tuning range in systems with fixed beam geometry. We note also that FPI of the pump components with their Stokes components can influence the dynamics of the nonlinear interactions when the medium is irradiated by several waves (or when several waves are produced in it). In particular, it explains the "lowering of the threshold" of the SRS, observed in gases and in liquids.[4,5]

The present paper is devoted to an exposition of the results of theoretical and experimental investigations of the dynamics of the generation of Stokes components in FPI, resulting from SRS of the additional pump or its two-photon absorption (TPA).

Principal attention is paid to the following questions:

a) The influence of the wave detuning on the generation efficiency.

b) The dependence of the dynamics of the process on the ratio of the frequencies of the strong and weak pump components.

c) Study of the singularities of the FPI in the case when the intensity of the weak pump is slightly lower than the SRS threshold.

d) Investigation of the possibility of conversion, into the low-frequency region, of radiation that is tunable in frequency or that has a broad spectrum.

II. GENERATION OF STOKES SRS COMPONENTS UNDER BIHARMONIC EXCITATION

1. Given-pump approximation

1.1. We shall investigate the interaction of waves in collinear beams, and consider therefore simultaneous interaction of fields $\mathbf{E}=\mathbf{e}_j[E(\omega_j)\exp(i\omega_j t)+\text{c. c.}]$, the frequencies of which are the condition

$$\omega_1-\omega_{1S}=\omega_2-\omega_{2S}=\omega_{21}. \quad (1)$$

The equations for the complex field amplitudes are

$$dC_1/dz=-\alpha_{11}C_1|C_{1S}|^2-\alpha_{12}C_2C_{2S}^*C_{1S}e^{i(\delta k)z}, \tag{2a}$$

$$dC_{1S}/dz=\alpha_{21}C_2^*C_{2S}C_1e^{-i(\delta k)z}+\alpha_{22}C_{1S}|C_1|^2, \tag{2b}$$

$$dC_2/dz=-\beta_{11}C_2|C_{2S}|^2-\beta_{12}C_1C_{1S}^*C_{2S}e^{-i(\delta k)z}, \tag{2c}$$

$$dC_2/dz=\beta_{21}C_1^*C_{1S}C_2e^{i(\delta k)z}+\beta_{22}C_{2S}|C_2|^2, \tag{2d}$$

where

$$\alpha_{11}=\frac{2\pi\omega_1^2nN_0\Gamma}{k_1c^2\hbar}|\varkappa(\omega_1)|^2,\quad \alpha_{12}=\frac{2\pi\omega_1^2nN_0\Gamma}{k_1c^2\hbar}\varkappa^*(\omega_1)\varkappa(\omega_2),$$
$$\alpha_{21}=\frac{2\pi\omega_{1S}^2nN_0\Gamma}{k_{1S}c^2\hbar}\varkappa^*(\omega_1)\varkappa(\omega_2),\quad \alpha_{22}=\frac{2\pi\omega_{1S}^2nN_0\Gamma}{k_{1S}c^2\hbar}|\varkappa(\omega_2)|^2, \tag{3}$$

where $|\varkappa(\omega_1)|^2$ and $\varkappa^*(\omega_1)\varkappa(\omega_2)$ are the contractions of the tensors $\varkappa_{ab}^*(\omega_1)\varkappa_{cd}(\omega_1)$ and $\varkappa_{ab}^*(\omega_1)\varkappa_{cd}(\omega_2)$ with respect to the index pairs a, b, and c, d;

$$\varkappa_{ab}(\omega_1)=\frac{1}{\hbar}\sum_q\left(\frac{(d_a)_{1q}(d_b)_{q2}}{\omega_{q1}-\omega_1}+\frac{(d_b)_{1q}(d_a)_{q2}}{\omega_{q2}+\omega_1}\right) \tag{4}$$

is the scattering tensor; n is the difference of the populations of level 1 and 2; N_0 is the particle-number density, Γ is the reciprocal line width;

$$\delta k=(k_2-k_{2S})-(k_1-k_{1S}) \tag{5}$$

is the wave detuning; an expression for β_{ij} is obtained from (3) by making the substitution $k_1, \omega_1 \rightleftarrows k_2, \omega_2$. The coefficients α_{ij} and β_{ij} can be expressed also in terms of the nonlinear susceptibilities, for example,

$$\alpha_{12}=(2\pi\omega_1^2/k_1c^2)\,\mathrm{Im}\,\chi^{(3)}\quad(\omega_1=\omega_2-\omega_{2S}+\omega_{1S}).$$

1.2. Assume that the system is acted upon by a biharmonic pump with components C_1 and C_2. If $|C_{1S}|^2 \ll |C_1|^2$ and $|C_{2S}|^2 \ll |C_2|^2$, then the amplitudes of the biharmonic-pumping components can be regarded as given. We assume henceforth that $C_1|_{z=0}=C_{10}<C_2|_{z=0}=C_{20}$ and all C_1 are the weak pump components and C_2 the strong ones. For the Stokes components of the pumping doublet we obtain from expressions (2b) and (2d) in the approximation[1)] $C_{1,2}=\mathrm{const}$

$$C_{2S}=\left\{\frac{K_{11}+i\delta k/2-\lambda_2}{\lambda_1-\lambda_2}e^{\lambda_1z}+\frac{K_{11}+i\delta k/2-\lambda_1}{\lambda_2-\lambda_1}e^{\lambda_2z}\right\}C_{1S0}e^{-i\delta kz/2}$$
$$+\left\{\frac{K_{12}(e^{\lambda_1z}-e^{\lambda_2z})}{\lambda_1-\lambda_2}\right\}C_{2S0}e^{-i\delta kz/2}, \tag{6a}$$

$$C_{2S}=\left\{\frac{K_{21}(e^{\lambda_1z}-e^{\lambda_2z})}{\lambda_1-\lambda_2}\right\}C_{1S0}e^{i\delta kz/2}$$
$$+\left\{\frac{K_{22}-i\delta k/2-\lambda_2}{\lambda_1-\lambda_2}e^{\lambda_1z}+\frac{K_{22}-i\delta k/2-\lambda_1}{\lambda_2-\lambda_1}e^{\lambda_2z}\right\}C_{2S0}e^{i\delta kz/2}, \tag{6b}$$

where

$$\lambda_{1,2}={}^1/_2(K^+\pm R^+\pm iR^-),$$
$$R^\pm=2^{-1/2}(\{[(K^+)^2-(\delta k)^2]^2+4(\delta k)^2(K^-)^2\}^{1/2}\pm[(K^+)^2-(\delta k)^2])^{1/2}, \tag{7}$$

and

$$K^\pm=K_{11}\pm K_{22},\quad K_{11}=\alpha_{22}|C_{10}|^2,\quad K_{22}=\beta_{22}|C_{20}|^2,$$
$$K_{12}=\alpha_{21}|C_{20}||C_{10}|,\quad K_{21}=\beta_{21}|C_{10}||C_{20}| \tag{8}$$

are the coefficients of the Raman and parametric gains at the frequencies ω_{1S} and ω_{2S}.

We consider three limiting cases.

a) $\delta k=0$—linear synchronism. From (6) we obtain $\lambda_1=K^+$ and $\lambda_2=0$, i.e., the increment λ_1 is equal to the sum of the coefficients of the Raman gain of the Stokes component under the influence of each pump separately. At low intensities of the weak pump component, the amplification efficiency does not depend on the intensity of the weak wave and is determined by the intensity of the strong pump.[2]

b) $(K^+)^2 \ll (\delta k)^2$. From (6) and (7) we easily find that in this case

$$C_{1S}\approx C_{1S0}e^{K_{11}z}+\frac{K_{12}}{\delta k}\exp\left\{i\,\mathrm{arctg}\frac{\delta k}{K^-}\right\}(e^{(K_{22}-i\delta k)z}-e^{K_{11}z})C_{2S0}, \tag{9a}$$

$$C_{2S}\approx C_{1S0}\frac{K_{21}}{\delta k}\exp\left\{i\,\mathrm{arctg}\frac{\delta k}{K^-}\right\}(e^{K_{22}z}-e^{(K_{11}+\delta k)z})+C_{2S0}e^{K_{22}z}. \tag{9b}$$

With increasing wave detuning, the efficiency of the parametric interaction decreases in proportion to $(\delta k)^{-1}$. As $\delta k\to\infty$, the processes become "decoupled" and the amplification of the Stokes SRS component takes place independently, i.e., only as a result of the Raman interaction with the corresponding pump.

c) $(K^+)^2 \gg (\delta k)^2$. Expanding the radicands of (7) in a series and retaining the first terms, we obtain

$$\lambda_1\approx K^+ + i\frac{\delta k}{2}\frac{K^-}{K^+},\quad \lambda_2\approx\frac{K_{11}}{2K^+}\frac{(\delta k)^2}{(K^+)^2}-i\frac{\delta k}{2}\frac{K^-}{K^+}, \tag{10}$$

with $\mathrm{Re}\,\lambda_1 \gg \mathrm{Re}\,\lambda_2$. From (10) and (6) we can obtain expressions for the field amplitudes; the real amplitudes (and consequently also the field intensities) vary approximately the same way as $\delta k=0$. The condition $(K^+)^2 \gg (\delta k)^2$ is equivalent to the phase-locking condition $\alpha_{11}^2W^2 \gg (\delta k)^2$ (see Sec. 3 of[6]).

1.3. In the given-pump approximation, a solution analogous to (6) can be obtained also for the process of generation of an anti-Stokes SRS component of frequency ω_{1a}.[6,12] In this case the increment of the anti-Stokes wave of the weak pump is close to $K_{22}(\omega_2, \omega_{2S})-K_{11}(\omega_1, \omega_{1a})$ if the condition $[K_{22}(\omega_2, \omega_{2S})-K_{11}(\omega_1, \omega_{1a})]^2 \gg (\delta k)^2$ is satisfied. Thus, at $K_{22}\gg K_{11}(|C_{20}|^2 \gg |C_{10}|^2)$ the effectiveness of the growth of the Stokes and anti-Stokes components of the weak pump are approximately equal, and in the opposite case, inequality $K_{22}>K_{11}$ is weak, and the increment of the Stokes component exceeds the increment of the anti-Stokes wave.

1.4. As follows from Sec. 1.2, at $\delta k=0$ the Stokes components increase monotonically. At $\delta k\neq 0$ the plots of $C_{1S}(z)$ and $C_{2S}(z)$ on the initial section can have an oscillating character. In the case of large values of δk it is easily shown from (9) that for $C_{1S0}\approx C_{2S0}$ at $z\lesssim K_{22}^{-1}$ the dependence of C_{2S} and C_{1S} on z is described by an oscillating function; at $z\gtrsim K_{22}^{-1}$ the beats become smoothed out, and the fields C_{1S} and C_{2S} increase monotonically (see also[7]). From an analysis of the expression for the phase difference of the fields

$$\theta(z)=(\delta k)z+(\varphi_2-\varphi_{2S})-(\varphi_1-\varphi_{1S})\quad(C_j=|C_j|\exp(i\varphi_j)),$$

it follows that in the first region we have $\theta(z)\sim(\delta k)z$, and in the second region we have $\theta\approx\tan^{-1}(\delta k/K^{-1})$ (for $(K^+)^2\ll(\delta k)^2$) or $\theta\approx\tan^{-1}(\delta k/K^+)$ (for $(K^+)^2\gg(\delta k)^2$). The transition of the $\theta(z)$ plot from the region of rapid linear variation into the region of slow motion corresponds to the phenomenon of the field locking.[8,9] We note that if $|C_{1s0}| \ll |C_{2s0}|$, then the phase locking takes place practically at the input, at $z=0$.

2. Scattering of weak pump in the presence of strong pump

At $\varkappa(\omega_2)|C_2 C_{2s}^*| \gg \varkappa(\omega_1)|C_1 C_{1s}^*|$ we can neglect the parametric terms in (2a) and (2b) and the Raman terms in (2c) and (2d). This approximation describes the dynamics of the interaction at $K_{22} \gg K_{11}$, so that $K^+ \approx K^- \approx K_{22}$ also for the phase-locking region (see Sec. 1.4). The equations for the field amplitudes C_{1s} and C_1 are easily obtained from (2):

$$\dot{C}_1 = -\gamma_1 e^{i\delta\xi} f(\xi) C_{1s}, \quad \dot{C}_{1s} = \gamma_2 e^{-i\delta\xi} f(\xi) C_1, \tag{11}$$

where the dot denotes the derivative with respect to ξ,

$$\delta = \frac{\delta k}{K_{22}}, \quad \gamma_1 = \frac{\omega_1^2}{\omega_{2s}^2}\frac{k_{2s}}{k_1}\frac{\varkappa(\omega_1)}{\varkappa(\omega_2)}\left|\frac{C_{2s0}}{C_{20}}\right|, \quad \gamma_2 = \frac{\omega_{1s}^2}{\omega_1^2}\frac{k_1}{k_{1s}}\gamma_1,$$
$$\xi = K_{22} z, \quad f(\xi) = e^{\xi}(1+b^2 e^{2\xi})^{-1}, \quad b^2 = \left|\frac{C_{2s0}}{C_{20}}\right|^2 \frac{\omega_2^2}{\omega_{2s}^2}\frac{k_{2s}}{k_2}. \tag{12}$$

2.1. *Case of zero wave detuning* $\delta k=0$. Solving the system of differential equations (11) we obtain

$$C_{1s} = C_{1s0}\cos\mathcal{A} + \frac{\omega_{1s}}{\omega_1}\frac{k_1^{1/2}}{k_{1s}^{1/2}} C_{10}\sin\mathcal{A},$$
$$C_1 = -\frac{\omega_1}{\omega_{1s}}\frac{k_{1s}^{1/2}}{k_1^{1/2}} C_{1s0}\sin\mathcal{A} + C_{10}\cos\mathcal{A}, \tag{13}$$

where

$$\mathcal{A} = R\,\mathrm{arctg}\, b(e^{\xi}-1), \quad R = \frac{\omega_{1s}\omega_1}{\omega_{2s}\omega_2}\left(\frac{k_{2s}k_2}{k_{1s}k_1}\right)^{1/2}\frac{\varkappa(\omega_1)}{\varkappa(\omega_2)}. \tag{14}$$

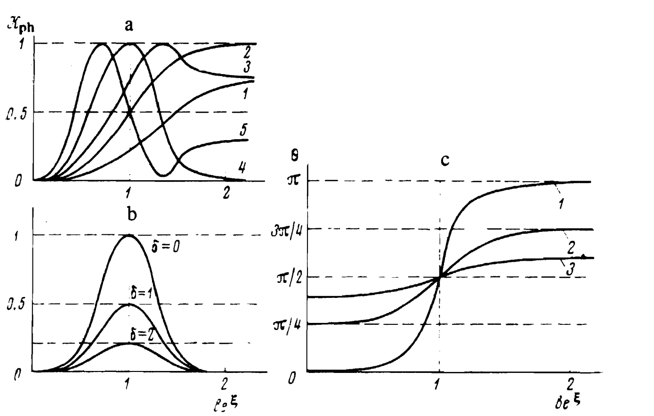

FIG. 1. Scattering of weak pump in the presence of strong pump: a) $\delta=0$, curves 1, 2, 3, 4, 5 correspond to plots of the proton coefficient of the conversion of the weak pump on the dimensionless coordinate ξ (the intensity of the strong pump at a fixed length of the nonlinear medium) respectively for $R<1$, $R=1$, $1<R<2$, $R=2$, $2<R<3$; b) dependence of the effectiveness of the interaction in the relative wave detuning $\delta = \delta k/K_{22}(R=2)$; c) dependence of the phase difference $\theta = (\delta k)z + (\varphi_2 - \varphi_{2c}) - (\varphi_1 - \varphi_{1c})$ on the dimensionless coordinate ξ at different values of the relative wave detuning δ; curves 1, 2, 3 correspond to the values $\delta_1 \ll 1$, $\delta_2 > \delta_1$, and $\delta_3 > \delta_2$.

If $R \approx 1$ (for example, at $\varkappa(\omega_1) \approx \varkappa(\omega_2)$ this corresponds to the case $\omega_1 \approx \omega_2$), then it follows from (13) that in the region $z \gtrsim K_{22}^{-1}$ the growth rates of the Stokes components C_{1s} and C_{12s} coincide. The asymptotic ($z \to \infty$) conversion of the weak pumping into the Stokes component was 100% in terms of the photons (see curve 2 of Fig. 1a).

At $R<1(\omega_1<\omega_2)$, the effectiveness of the transfer of the weak component is less than that of the strong one (see curve 1 of Fig. 1a). The limit of the conversion coefficient in terms of photons is

$$\mathcal{K}_{ph} = \lim_{z\to\infty}\frac{|C_{1s}|^2\omega_1^2 k_{1s}}{|C_{10}|^2\omega_{1s}^2 k_1} = \sin^2\left(R\frac{\pi}{2}\right). \tag{15}$$

At $R>1(\omega_1>\omega_2)$ the weak pump is converted more rapidly into its Stokes component that the strong pump. At

$$z_{max} = \frac{1}{K_{22}}\ln\left(\frac{1}{b}\,\mathrm{tg}\,\frac{\pi}{2R}\right) \tag{16}$$

the intensity of the Stokes component has a maximum $\mathcal{K}_{ph,max} = 100\%$ (Fig. 1a, curves 3–5).

2.2. *Case of arbitrary wave detuning* δk. From (11) we easily obtain two differential second-order equations for the complex amplitudes C_{1s} and C_1:

$$\ddot{C}_{1s} + \left(i\delta - \frac{1-b^2e^{2\xi}}{1+b^2e^{2\xi}}\right)\dot{C}_{1s} + R^2\frac{b^2e^{2\xi}}{(1+b^2e^{2\xi})^2}C_{1s} = 0,$$
$$\ddot{C}_1 - \left(i\delta + \frac{1-b^2e^{2\xi}}{1+b^2e^{2\xi}}\right)\dot{C}_1 + R^2\frac{b^2e^{2\xi}}{(1+b^2e^{2\xi})^2}C_1 = 0. \tag{17}$$

It is impossible to solve (17) in the general case. To assess the role of the wave detuning we consider the particular case $R=2$, when Eqs. (17) are ordinary differential equations. The solution of (17) at $C_{1s0} \approx 0$ takes the form

$$C_{1s} \approx \frac{\omega_{1s}}{\omega_1}\left(\frac{k_1}{k_{1s}}\right)^{1/2}\frac{C_{10}}{(1+\delta^2)^{1/2}}2be^{\xi}\exp\{-i(\delta\xi - \mathrm{arctg}\,\delta)\},$$
$$C_1 \approx C_{10}\frac{\{[1+\delta^2-b^2e^{2\xi}(1-\delta^2)]^2+4\delta^2b^4e^{4\xi}\}^{1/2}}{(1+b^2e^{2\xi})(1+\delta^2)} \times\exp\left\{i\,\mathrm{arctg}\frac{-2\delta^2e^{2\xi}}{(1+\delta^2)-b^2e^{2\xi}(1-\delta^2)}\right\}. \tag{18}$$

It follows therefore that in the presence of wave detuning the effectiveness of the parametric energy conversion into the Stokes component of the weak pump decreases like $[1+\delta^2]^{-1}$. The maximum conversion, equal to $[1+\delta^2]^{-1}\cdot 100\%$ in terms of photons, is reached at the same point as at $\delta k=0$ (see Fig. 1b). If $\delta^2 \ll 1$, then the conversion effectiveness remains practically the same as in the case of total synchronism ($\delta=0$). Such a weak dependence of the parametric interaction on the satisfaction of the conditions of the linear synchronism is connected with the phase locking phenomenon. To illustrate this, we obtain from (18) the phase difference θ:

$$\theta = \delta\xi - (\varphi_1 - \varphi_{1s}) + (\varphi_2 - \varphi_{2s}) = \mathrm{arctg}\,\delta + \mathrm{arctg}\frac{2\delta b^2e^{2\xi}}{1+\delta^2-b^2e^{2\xi}(1-\delta^2)}. \tag{19}$$

The dependence of θ on the coordinate ξ at different δ is shown in Fig. 1c. At $\delta \ll 1$ (curve 1) in the given-pump region, a phase difference $\theta_0 \approx 0$ is established, and the behavior of the system is analogous to its behavior in the case of total synchronism. With increasing δ, we have $\theta_0 \to \pi/2$ (curves 2 and 3 of Fig. 1c), and the effectiveness of the parametric interaction decreases; the coherent interaction of the field stops, and they vary only as a result of the Raman process.

3. Influence of TPA of the strong pump on the SRS of the weak pump

3.1. We consider the generation of the Stokes component of weak pump with frequency ω_1 in the case of TPA of the strong pump with frequency ω_2:

$$2\omega_2=\omega_1-\omega_{1s}=\omega_{21}. \tag{20}$$

The equations describing this process are analogous to Eqs. (2):

$$\begin{aligned} dC_1/dz&=-\alpha_{11}C_1|C_{1s}|^2-\alpha_{12}C_2^2C_{1s}e^{i(\delta k)z},\\ dC_{1s}/dz&=\alpha_{21}C_2^{*2}C_1e^{-i(\delta k)z}+\alpha_{22}C_{1s}|C_1|^2,\\ dC_2/dz&=-\beta_{11}|C_2|^2C_2-\beta_{12}C_1C_{1s}^*C_2^*e^{-i(\delta k)z}, \end{aligned} \tag{21}$$

where the notation of (3) is used, and $(\delta k)=k_1-k_{1S}-2k_2$.

At $\varkappa(\omega_2)|C_2|^2 \gg \varkappa(\omega_1)|C_1C_{1S}^*|$, the Raman conversion $C_1 \to C_{1S}$ and the variation of C_2 due to the parametric interaction can be neglected. We then obtain from (21)

$$\dot{C}_1=-\gamma_1\frac{e^{i\delta\xi}}{1+2\xi}C_{1s},\quad \dot{C}_{1s}=\gamma_2\frac{e^{-i\delta\xi}}{1+2\xi}C_1. \tag{22}$$

At zero detuning, the solution of the system (22) takes the form

$$\begin{aligned} C_{1s}&=C_{1s0}\cos\mathcal{L}+C_{10}\left(\frac{k_1}{k_{1s}}\right)^{1/2}\frac{\omega_{1s}}{\omega_1}\sin\mathcal{L},\\ C_1&=-C_{1s0}\frac{\omega_1}{\omega_{1s}}\left(\frac{k_{1c}}{k_1}\right)^{1/2}\sin\mathcal{L}+C_{10}\cos\mathcal{L},\\ \mathcal{L}&=\sqrt{\gamma_1\gamma_2}\ln\sqrt{1+2\xi}. \end{aligned} \tag{23}$$

The solutions (23) are approximately valid so long as the strong pump is not absorbed to a value on the order of the limiting value of the weak pump. If $|C_{1S0}| \ll |C_{10}|$ then total conversion of the weak pump into the Stokes component takes place over a length

$$z_{max}=\frac{1}{2\beta_{22}|C_{20}|^2}\left(\exp\frac{\pi}{\sqrt{\gamma_1\gamma_2}}-1\right). \tag{24}$$

It follows from (24) that at

$$\sqrt{\gamma_1\gamma_2}=\frac{\omega_1\omega_{1s}}{\omega_2^2}\frac{k_2}{\sqrt{k_1k_{1s}}}\frac{\varkappa(\omega_1)}{\varkappa(\omega_2)}=\pi$$

to ensure 100% conversion under the conditions of linear synchronism ($\delta k=0$) an absorption of the strong pumping by a factor e should take place over length L of the active medium.

3.2. It is impossible to solve the system (21) at an arbitrary ratio of the field intensities and at $\delta k \neq 0$. However, at $|C_{10}||C_{20}| \gg |C_{1S0}|^2$ an approximate solution of (21) can be constructed using the equations for the real slow amplitudes $A_j=C_j\exp[-i\varphi_j(z)]$ and the phase difference $\theta=-2\varphi_2+\varphi_1-\varphi_{1S}+\delta kz$, which are given by

$$\begin{aligned} \frac{dA_1}{d\xi_1}&=-A_{1s}(A_1A_{1s}+sA_2^2\cos\theta),\quad \frac{dA_{1s}}{d\xi_1}=q_{1s}A_1(A_1A_{1s}+sA_2^2\cos\theta),\\ \frac{dA_2}{d\xi_1}&=q_2A_2(sA_2^2+A_1A_{1s}\cos\theta),\quad \frac{d\theta}{d\xi_1}=\frac{\delta k}{\alpha_{11}}+W(A_1,A_{1s},A_2)\sin\theta, \end{aligned} \tag{25}$$

where

$$W(A_1,A_{1s},A_2)=2q_2A_1A_{1s}+sA_2^2\left(\frac{A_{1s}}{A_1}-q_{1s}\frac{A_1}{A_{1s}}\right), \tag{26}$$

and

$$\xi_1=\alpha_{11}z,\quad s=\frac{\varkappa(\omega_2)}{\varkappa(\omega_1)}\quad q_{1s}=\frac{\omega_{1s}^2}{\omega_1^2}\frac{k_1}{k_{1s}},\quad q_2=\frac{\omega_2^2}{\omega_1^2}\frac{k_1}{k_2}s$$

Inasmuch as $A_{1S0} \ll A_{10}$ and $A_{1S0} \ll A_{20}$, it follows that in the initial region of the interaction we have $|W\alpha_{11}|^2 \gg (\delta k)^2$. As shown in[10,11], in this case phase locking of the fields takes place, at which $\theta \approx \delta k/\alpha_{11}W$ (it is easy to establish from (26) that in the given-pump approximation the condition $(\delta k)^2 \ll \alpha_{11}^2W^2$ is equivalent to the condition $\delta^2 \ll 1$, see (12)). The coefficient W decreases with changing fields; in the region $W(A_1,A_{1S},A_2) > \delta k/\alpha_{11}$ we can put $\theta \approx 0$.[11] The expression obtained in this manner from (25) for the conversion length to $I_1 \to I_{1S}(I_j=A_j^2)$ takes the form

$$L=\frac{1}{2\alpha_{11}}\int\limits_{I_{1s0}}^{I_{1s}}\frac{dI_{1s}}{I_{1s}(M_1-I_{1s})+sI_2[q_{1s}I_{1s}(M_1-I_{1s})]^{1/2}}, \tag{27}$$

where

$$I_2=I_{20}\exp\left\{\frac{2q_2}{q_{1s}}\left[\arcsin\left(\frac{I_{1s0}}{M_1}\right)^{1/2}-\arcsin\left(\frac{I_{1s}}{M_1}\right)^{1/2}\right]\right\}, \tag{28}$$

$$M_1=q_{1s}I_1+I_{1s}=q_{1s}I_{10}+I_{1s0}. \tag{29}$$

The solution (27) is valid in the region of values satisfying the inequality

$$|W|=\left|\frac{2q_2}{q_{1s}}[q_{1s}I_{1s}(M_1-I_{1s})]^{1/2}+sI_2\frac{2I_{1s}-M_1}{[q_{1s}I_{1s}(M_1-I_{1s})]^{1/2}}\right|>|\delta k|, \tag{30}$$

and for arbitrary ratios I_{10}/I_{20} and $\delta k/K_{22}I_{20}$.

3.3. By way of an example, let us estimate the possibility of obtaining radiation in potassium vapor by generating a Stokes SRS component from a low-power dye laser that is tunable in the range 410–460 nm, see Fig. 2 (the Stokes component will be tunable in the range 3–15 μ), in the presence of TPA of the radiation of a high-power source with $\lambda=951$ nm. The quantities $\varkappa(\omega_2)$ and $\varkappa(\omega_1)$, calculated in the approximation that the levels $4P$ and $5P$ take part in the two-photon transitions, are respectively equal to 2.2×10^{-23} and 1.2×10^{-22} cm^3 (for $\lambda_1=455$ nm and $\lambda_{1S}=9$ μ). At a pressure of 15 torr, $\alpha_{11}=2\times10^{-5}$ cgs esu $I_{20}=2.5\times10^8$ W/cm^2, $I_{10}=2.5\times10^7$ W/cm^2, and $I_{1S0}=10^{-6}I_{10}$ we have $\delta k \approx 1.25$ cm, and the Raman-parametric generation in the region (30) ensures an $I_1 \to I_{1S}$ conversion coefficient ~30% in terms of photons,[3)] the conversion length amounting to 15 cm.

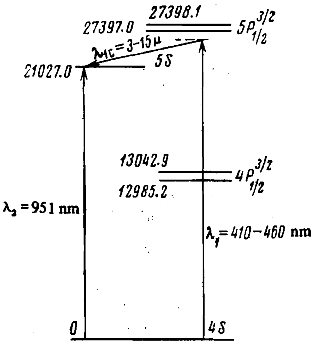

FIG. 2. Generation of weak-pump SRS Stokes component in the presence of strong-pump TPA in KI vapor.

In the case of confocal focusing the laser powers needed to realize the process are $I_{20}\approx 100$ kW and $I_{10}\approx 3$ kW. In the absence of the $\lambda=951$ nm radiation, to obtain an analogous conversion the required length of the active region of the medium is 600 cm (see the first term in the denominator of the integrand of (27)).

III. EXPERIMENTAL INVESTIGATION OF THE GENERATION OF STOKES COMPONENTS IN BIHARMONIC EXCITATION

1. We investigated the excitation of Stokes components of SRS in compressed hydrogen under the influence of isolated and biharmonic pumps. The strong pump I_2 was the second harmonic of a frequency-stabilized single-mode neodymium laser ($\lambda_2=530$ nm). The pump source I_1 was the Stokes SRS component ($\lambda_1=630$ nm) from the 530-nm radiation, formed in a chamber with deuterium (Fig. 3a). The 630-nm radiation and the unconverted part of the second harmonic were focused into a chamber with hydrogen (pressure 75 atm).

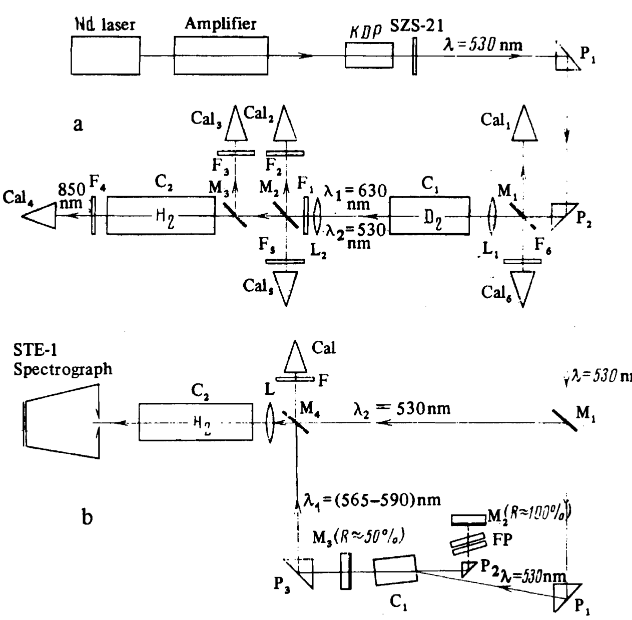

FIG. 3. Diagram of experimental setups for the investigation of SRS with biharmonic pumping. a) C_1—chamber with deuterium (length 16 cm, pressure 90 atm), C_2—chamber with hydrogen (length 44 cm, pressure 75 atm), M_1—M_3—beam-splitting glass plates, L_1—L_3—lenses, Cal_1—Cal_6—calorimeters, F_1—F_6—sets of absorbing and interference light filters; b) C_1—cell with rhodamine 6Zh solution in alcohol, C_2—chamber with hydrogen.

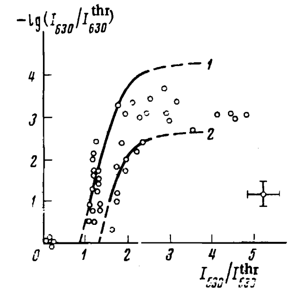

FIG. 4. Decrease of threshold intensity I_{10} as a function of the excess of the power of the strong pump I_{20} over the threshold SRS: curves 1 and 2 were calculated for the values $(I^r_S)_{min}$ equal to 0.1 and 5 W, respectively.

The ratio of the pump intensities I_{10} and I_{20} at the input was varied with light filters. The intensity of the Stokes component I_{1S} ($\lambda_{1S}=850$ nm) was registered with a calorimeter having a maximum sensitivity $I^r\approx 0.1$ W.

We measured the ratio $I_{10}(I^r)/I_{10}^{thr}$ (I_{10}^{thr} is the SRS threshold in the absence of the strong pumping I_{20}; $I_{10}(I^r)$ is the input intensity sufficient for the appearance of I_{1S} at a given receiver sensitivity I^r) as a function of the quantity I_{20}/I_{20}^{thr}. The SRS threshold was $I_{20}^{thr}=50$ kW at the wavelength λ_2 and $I_{10}^{thr}\approx 5$ kW at the wavelength λ_1.[4)] An increase of I_{20} to 150 kW led to the decrease of I_{10} needed to obtain $I_{1S}>0.1$ W by a factor 5×10^3 in comparison with I_{10}^{thr} ($I_{10}\sim 1$ W).

2. To compare the experimental results with theory, we calculated the dependences of the "lowering of the SRS threshold," characterized by the quantity I_{10}/I_{10}^{thr}, on the excess I_{20}/I_{20}^{thr} of the strong-pump power over the threshold value. The calculation was carried out in accordance with the results of Sec. II. The registered value of the Stokes component was $I^r_{1S}\lesssim(0.2-0.5)I_{10}$, and then, as follows from Figs. 1a and 1c, it is possible to put in this region $\theta=\theta_0=\tan^{-1}(\delta k/\beta_{22}I_{20})=\text{const}$. Inasmuch as in this region I_{10} cannot be regarded as specified, we solved the corresponding equations for real amplitudes and obtained the dependence of I_{10}/I_{10}^{thr} on I_{20}/I_{20}^{thr} for the given value of I^r_{1S}:

$$\frac{I_{10}}{I_{10}^{thr}}=\frac{\omega_1}{\omega_{1S}}\frac{I^r_{1S}}{I_{20}^{thr}}\sin^{-2}\{R\cos\theta_0 \operatorname{arc\,tg} be^{g_{22}I_{20}L}\}. \quad (31)$$

In accordance with the experimental conditions $(\Gamma N_0)=4.5\times 10^{11}$ cgs esu,[13] $\varkappa^2(\omega_1)=\varkappa^2(\omega_2)\approx 0.68\cdot 10^{-50}$ cgs esu,[12] $\delta k=5.6$ cm^{-1},[14] $L=5$ cm, $I_{10}^{thr}\approx 5.7\times 10^8$ W/cm^2, $I_{20}^{thr}\approx 4.4\times 10^8$ W/cm^2, $I_{1S0}\approx I_{2S0}=10^{-10}I_{10}, I_{20}$, $\theta_0\approx\pi/4$. The scatter of the energies of the pulses of the initial laser which leads also to a scatter of $I_{10}(\lambda=630$ nm) was such that the signal registered at $\lambda_{1S}=850$ nm ranged from 0.1 to 5 W. The corresponding plots of $-\log(I_{10}/I_{10}^{thr})$ for the registered powers $I^r\approx 10^{-3}I_{20}^{thr}$ ($I^r\approx 5$ W) and $I^r\approx 2\times 10^{-5}I_{20}^{thr}$ ($I^r\approx 0.1$ W) are shown in Fig. 4. The experimental points of I_{10}/I_{10}^{thr} and I_{20}/I_{20}^{thr} lie between the calculated curves.[5)] In the investigated case we have $R=0.8$ (see (14)) and the asymptotic conversion in terms of photons, due to the parametric interaction, is $\mathscr{K}_{ph}\approx 0.9$ (see (15), Fig. 1a). The maximum conversion coefficient obtained in the experiment was ~50%.

3. To verify that the appearance of the Stokes component I_{1S} of the weak pump I_1 in the presence of the strong pump I_2 is due to a parametric process, an experiment was set up in which waves with λ_1 and λ_2 were directed in the chamber opposite to each other. Then $|\delta k|^{-1} \approx \lambda_1$ and no locking is possible. To this end, the weak pump I_1 was the backward Stokes SRS radiation from $\lambda_2 = 530$ nm into H_2 ($\lambda_1 = 680$ nm), and the strong pump was as before, the second harmonic of a neodymium laser $\lambda_2 = 530$ nm. At the same values of I_{10} as in the scheme with the same direction of pump propagation, the Stokes component I_{1S} was not observed even at $I_{20}/I_{20}^{\text{thr}} \approx 30$.

4. As follows from the theoretical analysis, the dependence of the Stokes radiation at the frequency ω_{1S} on the coordinate (or on the power of the strong pump at a fixed cell length), can be nonmonotonic if, for example, $R > 1$ (Fig. 1a) (in the case of hydrogen this corresponds to the case when the strong pump is of the lower frequency). Then the dependence of the power of the Stokes component I_{1S} on the intensity I_2 at a fixed value of the weak pump I_1 has a sharp maximum.

To verify this, we performed an experiment in which the strong and weak pumps were the radiation of the first and second harmonics of a neodymium laser, $\lambda_1 = 530$ and $\lambda_2 = 1060$ nm. The radiation at the Stokes frequencies $\gamma_{1S} = 680$ and $\lambda_{2S} = 1880$ nm and at the anti-Stokes frequency $\lambda_{1a} \approx 434$ nm were registered with calorimeters. The SRS thresholds were $I_{10}^{\text{thr}} \approx 30$ kW and $I_{20}^{\text{thr}} = 250$ kW, the length of the region of the effective interaction was $L \approx 10$ cm, and the wave detuning was $\delta k \approx 9.6$ cm.[14] The weak pump was varied in the range $(0.4\text{–}0.6) I_{10}^{\text{thr}}$. The experimental results are shown in Fig. 5. The statistical picture of many flashes, shown in Fig. 5, shows reliably the presence of a rather sharp maximum in the region of the intensity of the strong pump $I_{20} \approx 2 I_{20}^{\text{thr}}$, where its coefficient of conversion into the Stokes component is $\mathscr{K}_{\text{ph}2S} \approx (20\text{–}35)\%$ (in terms of photons). The theoretical curve calculated for these conditions and $I_{10} \approx 0.5 I_{10}^{\text{thr}}$ by means of formulas (13) (i.e., without allowance for the Raman interaction of the weak pump) is also shown in Fig. 5 (curve 2). The position of the maximum on this curve corresponds to $\mathscr{K}_{\text{ph}2S} \approx 28\%$, which is close to the experimentally observed position.

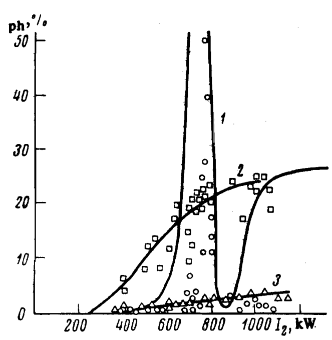

FIG. 5. Dependence of the conversion coefficients in terms of photons, $\mathscr{K}_{\text{ph}}$ for the Stokes components of the strong I_{2S} (curve 2) and weak I_{1S} (curve 1—calculated) pumps and of the anti-Stokes component (curve 3) of the weak pump vs the power I_2 of the strong pump; $I_{10} = (0.4 - 0.6) \cdot I_{10}^{\text{thr}}$, $\Delta - I_{1a}$, $\circ - I_{1S}$, $\square - I_{2S}$.

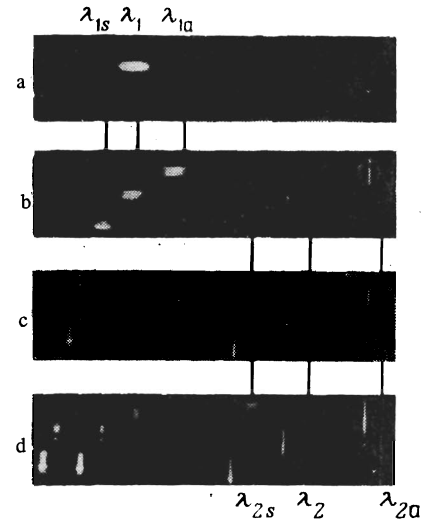

FIG. 6. Spectrograms of the radiation at the exit from the chamber with hydrogen exposed to the second harmonic of a neodymium laser and a broad-band or narrow-band tunable rhodamine laser. The explanation is in the text (see Sec. III).

The very presence of a sharp maximum indicates that the FPI plays a decisive role in the generation of the Stokes component of the weak pump even at $I_{10} \approx 0.5 I_{10}^{\text{thr}}$. At these values of I_{10}, the behavior of the anti-Stokes component I_a differs significantly from that of the Stokes component (curve 3 of Fig. 5), and in the same interval of the intensities I_{20} the anti-Stokes component is much weaker than the Stokes component, which is in agreement with the results of the theoretical analysis (see Sec. II, subsection 1). It appears that this is aided also by the six-photon interaction with simultaneous participation of the Stokes and anti-Stokes waves from the weak pump. It is qualitatively clear that the latter should lead to an even larger decrease of the growth rate of the anti-Stokes component in comparison with the Stokes component, owing to the transfer of the energy I_{10} into I_{1S}.

5. The processes described above are not very critical to the wave detuning, so that it is possible to obtain conversion of broad-band or tunable weak pumping. We have performed an experiment on the conversion of dye-laser radiation into the Stokes and anti-Stokes regions. The experimental setup is shown in Fig. 3b. The dye-laser radiation (rhodamine 6Zh goes directly together with the second harmonic of the neodymium laser into a chamber with hydrogen. The spectra at the exit from the chamber were registered with an STÉ-1 spectrograph and are shown in Fig. 6. Figures 6a and 6b show spectrograms obtained respectively in the absence and in the presence of SRS of the strong pump. In the absence of the $\lambda_2 = 530$ nm radiation, the threshold of the SRS at $\lambda_1 \approx 575$ nm ($\Delta\lambda \approx 20$ nm) was ~ 500 kW. The spectra in Figs. 6a and 6b were obtained in $P_1 \sim 10$ kW, $P_2 \approx 500$ kW ($P_2^{\text{thr}} \approx 50$ kW).

Figure 6c shows the spectrum at the exit of the chamber with hydrogen at a radiation with $\Delta\lambda \approx 2$ nm ($P_1 \approx 10$

kW), while Fig. 6d shows the same but with the dye-laser wavelength tuned with the aid of a Fabry–Perot resonator. When the frequency of the rhodamine laser was varied in the range 562–585 nm, the anti-Stokes and Stokes radiation was tuned in the ranges 455–469 and 737–773 nm. As follows from the spectrograms, the efficiency of the conversion of the weak pumping is of the order of 50% in energy.

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1)The increments $\lambda_{1,2}$ were given in[2] for the case $K_{22} \gg K_{11}$.
2)Expressions for C_{2S} and C_{1a}^* can be easily obtained from (6) and (7) by making the substitutions $K_{11} \to K_{11}(\omega_1, \omega_{1a})$, $K_{12} \to -K_{12}(\omega_2, \omega_{1a})$ and $C_{1a} \to C_{1a}^*$.
3)The solution of the transcendental equation (30) and the calculation of the integral (27) were carried out with a computer.
4)The difference between the threshold intensities cannot be explained by the frequency dependence of the cross sections of the spontaneous Raman scattering, and is apparently connected with the somewhat different geometry of the beams of the slow and weak pumps under the experimental conditions.
5)Curves 1 and 2 (Fig. 4) were obtained in[1] by an approximate calculation in the phase-locking regions (see Sec. III, subsection 3), i.e., under the assumption $\theta \approx 0$ and $\delta \ll 1$.

1G. V. Venkin, G. M. Krochik, L. L. Kulyuk, D. I. Maleev, and Yu. G. Khronopulo, Pis'ma Zh. Eksp. Teor. Fiz. 21, 235 (1975) [JETP Lett. 21, 105 (1975)].
2J. A. Giordmaine and W. Kaiser, Phys. Rev. 144, 676 (1966).
3S. A. Akhmanov and N. I. Koroteev, Zh. Eksp. Teor. Fiz. 67, 1306 (1974) [Sov. Phys. JETP 40, 650 (1975)].
4J. A. Duardo, F. M. Johnson, and L. J. Nugent, IEEE J. Quantum Electron. 4, 397 (1968).
5Ya. S. Bobovich and A. V. Bortkevich, Opt. Spektrosk. 31, 424 (1971).
6G. M. Krochik and Yu. G. Khronopulo, Kvantovaya Elektron. 2, 1693 (1975) [Sov. J. Quantum Electron. 5, 917 (1975)].
7D. N. Klyshko, Zh. Eksp. Teor. Fiz. 64, 1160 (1973) [Sov. Phys. JETP 37, 590 (1973)].
8S. A. Akhmanov, V. G. Dmitriev, V. P. Modenov, and V. V. Fadeev, Radiotekh. Élektron. 10, 2157 (1965).
9V. S. Butylkin, G. V. Venkin, V. P. Protasov, N. D. Smirnov, Yu. G. Khronopulo, and M. F. Shalyaev, Pis'ma Zh. Eksp. Teor. Fiz. 17, 400 (1973) [JETP Lett. 17, 285 (1973)].
10G. L. Gurevich and Yu. G. Khronopulo, Zh. Eksp. Teor. Fiz. 51, 1499 (1966) [Sov. Phys. JETP 24, 1012 (1967)].
11V. S. Butylkin, G. M. Kronik, and Yu. G. Khronopulo, *ibid.* 68, 506 (1975) [41, 147 (1975)].
12A. G. Ford and J. C. Browne, Atomic Data 5, 305 (1973).
13P. Lallemand, P. Simowa, and G. Brett, Phys. Rev. Lett. 17, 1239 (1966).
14M. Born and E. Wolf, Principles of Optics, Pergamon, 1970.

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Resonance two-electron charge exchange

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The process of resonance exchange of two electrons in an atom–ion collision is considered. At large internuclear distances the process proceeds mainly as a superposition of two inelastic transitions for each separate electron. A substantial contribution to the charge exchange is also made by parallel electron transitions in which the energy of each electron does not change. For the purpose of computing the probability of these transitions, the asymptotic behavior of the wave function resulting from the simultaneous removal of two electrons from the atom is investigated.

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1. The probability of resonance charge exchange of any number of electrons in a collision between an atom and its multiply charged ion can be computed if the spacing between the energy terms of the even and odd—with respect to the interchange of the nuclei—states is known.[1] At collision velocities lower than the orbital velocities of the bound electrons, the transition occurs at large interatomic distances, so that it is necessary to know the asymptotically exact value of the term spacing. This spacing exponentially decreases as the atoms move away from each other.

The term spacing for a two-electron exchange was earlier computed in the papers[2,3] and estimated in[4]. In[2] the computation was carried out, using unperturbed atomic wave functions, i.e., according to the Heitler-London scheme. In[3,4] it is pointed out that, in order to obtain the asymptotic form of the term splitting for large internuclear distances, it is necessary to construct the correct wave function of the outer atomic electron in the vicinity of the perturbing ion.

In[3,4] the contribution made by the crossover transitions to the splitting is investigated: An electron from the outer (inner) orbit of the atom a crosses to an inner (outer) orbit of the atom b. These transitions are inelastic for each separate electron (the transition of both electrons as a whole is elastic). The dominant contribution to the transition probability is made here by the configuration in which the electrons move apart on different nuclei, and the independent electron approximation as a zeroth approximation is valid.[3,5–7] Inelastic

