Investigation of the four-photon resonance scattering of light

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The frequency-angular diffusion of radiation from a dye laser propagating in potassium vapor (in the neighborhood of the D lines) in the presence of intense resonance radiation is investigated experimentally. An increase in the divergence of the radiation from a dye laser (DL) and a broadening of the absorption lines (D_1 and D_2) and of the reversed Raman scattering line is observed in the case of unidirectional propagation. These effects are absent when the intense radiation and DL radiation are propagated in opposite directions. The results obtained are interpreted as a consequence of four-photon resonance parametric scattering.

1. INTRODUCTION

A broadening of the spectrum of radiation passing through a resonance medium (potassium vapor) was first observed by Movsesyan et al.[1] Later a number of authors investigated both the broadening of the spectrum and a change in the divergence of the intensity of the radiation in resonance atomic media[2,3]. In our work[2] these phenomena were related to the four-photon resonance parametric scattering (FRPS). The processes mentioned above require phase synchronization both in space and in time which is violated in the case of scattering of weak waves which are propagated in the opposite direction to a strong field. Experiments carried out with oppositely directed beams[4,5] have indeed shown a sharp decrease in the broadening of the spectrum of a weak oppositely directed wave and the absence in it of an increase of angular divergence. In the work of Bonch-Bruevich, Khodovoĭ, and Khromov[6] a weak "test" field was directed at an angle θ_1 to the beam of an intense field and the appearance of an additional beam at an angle $\theta_1 = -\theta_1$ was observed, this being also related to FRPS.

Discussions were given in the literature also of other interpretations of the phenomena indicated above. Thus, in[3,6] it was assumed that as a result of the nonlinear dependence of the index of refraction on the field amplitude under resonance conditions self-focusing or self-defocusing of light occurs and it is this that leads to an increase in the spatial divergence of the radiation. The self-focusing observed under closely similar conditions[3-7] speaks in favor of such an explanation, and this is also supported by the disappearance of angular divergence in the case of a formation of a beam of relatively large diameter and sufficiently homogeneous over its cross section.[6] A broadening of the spectrum was interpreted in[3] as the appearance of phase modulation of light as a result of amplitude modulation (AM-PM conversion)[8].

In the present paper we describe experiments the results of which indicate, in our opinion, the essential role played by the FRPS in the case of propagation of intense radiation in a resonant medium (potassium vapor) and the inadequacy of an explanation based on self-focusing and AM-PM conversion.

2. EXPERIMENTAL SETUP AND CONDITIONS

A diagram of the experimental arrangement is shown in Fig. 1. Potassium vapor filling the cell 7 (cell length

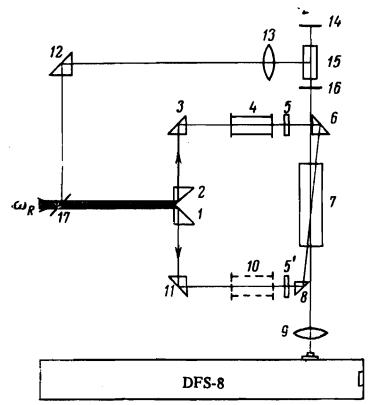

FIG. 1. Arrangement of the experimental apparatus (for notation cf., text).

15 cm) was used as the resonant medium. The interacting beams were provided by:

1) Stokes stimulated Raman scattering (SRS) of the radiation from a ruby laser in nitrobenzene ($\omega_S = 13055\ cm^{-1}$, $\Delta\omega_S = 4\ cm^{-1}$, $P_{S\,max} = 5\ MW/cm^2$).

2) Radiation from a dye laser (DL) excited by part of the radiation from a ruby laser ($\omega_R = 14400\ cm^{-1}$, $\Delta\omega_R = 0.02\ cm^{-1}$, $P_{R\,max} = 50\ MW/cm^2$).

The spectrum of the DL radiation had a width of approximately 250 cm^{-1} with a central frequency $\omega_k = 13060\ cm^{-1}$, and its intensity was not great, but sufficient to record a spectrogram during one pulse. Observations were carried out in the region of the resonance doublet of potassium $4S_{1/2} - 4P_{1/2}$ ($\omega_1 = 12985.2\ cm^{-1}$) and $4S_{1/2} - 4P_{3/2}$ ($\omega_2 = 13042.9\ cm^{-1}$).

For the excitation of the DL (a solution of a brilliant green dye in glycerine) a portion of the radiation from the ruby laser was split off by the light partitioning plate 17 and was focused by the cylinderical lens 13 (f = 9 cm) within the cell 15 containing the dye. The mirror 14 (R = 95%) and the glass plate 16 form the resonator for the DL. The DL radiation propagated along the axis of the cell containing the potassium and was focused by the lens 9 (f = 24 cm) on the slit of the DFS-8 spectrograph in such a manner that it would be situated in the focal plane of the lens 9. As a result the spectrogram recorded both the spectrum and the angular divergence of the radiation being investigated (the frequency-angular distribution).

The principal part of the ruby laser radiation was utilized for the excitation of SRS in nitrobenzene. For

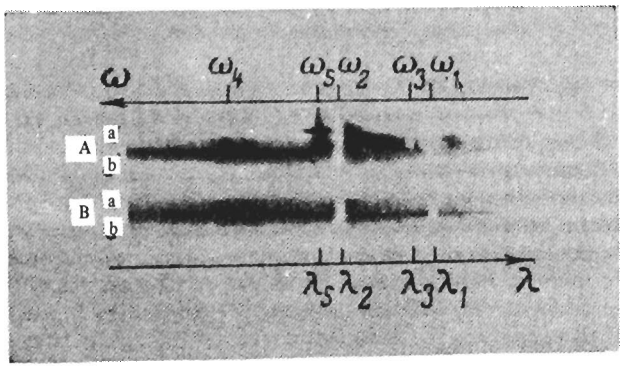

FIG. 2. Spectrum of the radiation from a dye laser in the region of the transitions $4S_{1/2}$-$4P_{3/2,1/2}$. The spectrograms correspond to the following conditions: Aa-intense field and $k_S \uparrow\uparrow k_K$, Ba strong field and $k_S \uparrow\downarrow k_K$; Ab, Bb are the DL comparison spectra (without the intense field). Vapor pressure p = 0.028 Torr.

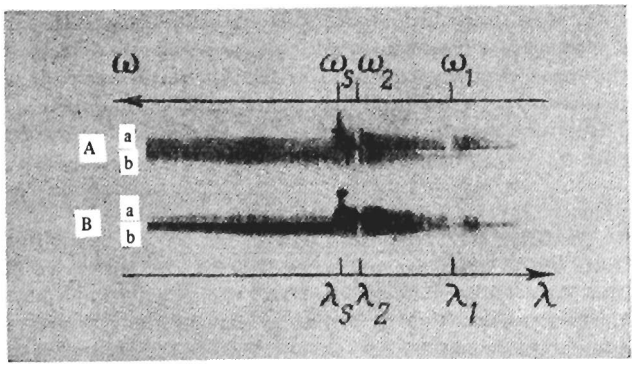

FIG. 3. The same as in Fig. 2, but at a pressure of p = 0.0038 Torr.

this the laser beam was directed by the deflecting prisms 2 and 3 into the cell 4 filled with nitrobenzene. The light filter 5 (FS-6) cut off the radiation from the laser and the beam of SRS was directed with the aid of the prism 6 into the cell 7 containing potassium vapor at a small angle (2°—3°) to its axis. In this case both beams k_S and k_K were propagated in approximately the same direction. The diameter of the SRS beam on entering the cell containing potassium vapor was equal to 10 mm. The cross section of the DL beam had a rectangular shape of width 2 mm and of height 5 mm (on entering the cell containing potassium).

To accomplish propagation of the SRS and DL beams in opposite directions ($k_S \uparrow\downarrow k_K$) the cell containing the nitrobenzene was placed in position 10, and the beam from the ruby laser followed the path 1-11-10-5. The SRS radiation followed the path 10-5-8-6 and in the cell 7 travelled along the same line as in the case $k_S \uparrow\uparrow k_K$, but in the opposite direction. The disposition of the apparatus was symmetric and the change in the direction of k_S did not lead to any appreciable change in the flux density of the SRS radiation in the cell 7. This was achieved by making the distances 2-3-4, 1-11-10 and 4-6-8, 10-8-6 equal and by choosing as far as possible identical elements in the two branches of the experimental arrangement (the filters 5,5, the prisms 1,2 and 11,3) etc.

The spectrum was recorded with the aid of the spectrograph DFS-8 with a diffraction grating of 1200 lines/mm, in first order, with a linear dispersion of 4.8 cm^{-1}/mm. For a comparison of the spectrum of the dye laser in the presence of the intense field (SRS) and in its absence a diaphragm was used situated at the entrance slit of the spectrograph: it covered in turn the upper and the lower (along the height of the slit) half of the DL beam; one half was used to photograph the DL spectrum when the SRS radiation passed through the potassium vapor, and the second half was used to photograph the DL spectrum in the absence of SRS (cf. Figs. 3 and 2 below). The photographs were made using "Infra-760" plates and an Abbé comparator was used to measure the spectrograms.

3. EXPERIMENTAL RESULTS

Figure 2Aa reproduces the spectrum of the radiation from a dye laser which has passed through the cell containing potassium vapor (pressure p = 0.028 Torr) simultaneously with the Stokes SRS radiation in nitrobenzene; the directions of propagation of the DL and the SRS radiation coincided ($k_S \uparrow\uparrow k_K$): the cell containing nitrobenzene was placed in position 4 (cf., Fig. 1), and the ruby laser beam traversed the path 2-3-4-5-6-7. Figure 2Ab represents a spectrogram of the DL radiation that has traversed the cell 7 containing potassium vapor in the absence of SRS, but in all others respects under the same conditions as for Fig. 2Aa: the path of the beam 2-3-4-5-6 was blocked by a nontransparent screen in front of the prisms 1,2. The spectrograms of Fig. 2Ba and Bb were obtained for the SRS and DL beams travelling in opposite directions, and in all other respects under the same conditions as the spectra of Fig. 2Aa and Ab. Finally, Fig. 3 corresponds to a smaller pressure (p = 0.004 Torr), while the letter portion of the labels refers the spectrograms to the same conditions as in the case of Fig. 2.

From a comparison of Fig. 2Aa and Fig. 2Ab it can be seen that the intense SRS field of frequency ω_S propagated in the same direction as the radiation from the dye laser DL alters in an essential manner the spectrum of the DL radiation. First of all, in the domain of frequencies adjacent to the lines of the resonance doublet ω_1 and ω_2 on the long wavelength side, the angular divergence of the DL radiation has been increased. Within these segments of the spectrum the frequency-angular diagram of the radiation has a characteristic "whisker" shape. An analogous diagram for the scattered radiation when the potassium vapor was irradiated only by the SRS beam has been observed by us previously[2].

The change in the widths of the lines ω_1 and ω_2 is noteworthy. In the absence of the intense field the width is $\Delta\omega_1 = 3$ cm^{-1}, while the intense field in the case $k_S \uparrow\uparrow k_K$ leads to its being increased to $\Delta\omega_1 \approx 9$ cm^{-1} (Fig. 2Aa and Ab). The width of the absorption line ω_2 (the transition $4S_{1/2} - 4P_{3/2}$) varies along the height of the spectrum, i.e., depends on the angle between the axis of the beam and the direction towards a given point of the slit. In the direction of the maximum intensity of the intense field (the central portion of the slit) the width has a minimum value (approximately 1.5 cm^{-1}), while in the region of diminishing intensity it increases monotonically (upwards in the spectrum of Fig. 2Aa). In Fig. 2A one can see a field shift of the line ω_2 in the red direction (approximately by 1.0 cm^{-1}). The line ω_1 is also shifted towards the long wavelength side, but it is difficult to estimate this shift (as a result of the large line width). The direction of the shift of the lines in Fig. 2Aa in comparison with Fig. 2Ab is in agreement with the concept of the splitting of the lines by the field[9-12].

In addition to the absorption lines ω_1 and ω_2 of the resonance doublet one can see in the spectrogram of Fig. 2Aa a new "absorption" line whose frequency ω_3 is shifted by 58 cm^{-1} with respect to the frequency ω_S of the SRS radiation. The width of this line is $\Delta\omega_3 \approx 5$ cm^{-1}. This shift of the line (with respect to ω_S) is equal to the fine structure splitting of the 4P term. Therefore we consider that the line ω_3 corresponds to the so-called inverse Raman scattering: a potassium atom in the $4P_{3/2}$ state emits a photon of the intense field ω_S, absorbs a photon ω_3 from the DL spectrum and finds itself in the $4P_{1/2}$ state. As far as we know, this is the first observation of inverse Raman scattering in atomic vapors. In the case of potassium under closely similar conditions usually a Raman scattering line of the intense field appears[13-15]: a photon ω_S disappears, a photon of frequency $\omega_4 = \omega_S + 58$ cm^{-1} is emitted which is symmetric to the frequency ω_3 with respect to ω_S, and the atom goes over from the $4P_{3/2}$ state into the $4P_{1/2}$ state. In Fig. 2Aa increased blackening is observed in this region of the spectrum, but the structure of the spectrum is too diffuse for us to be able to ascribe confidently this blackening to Raman scattering.

The changes in the DL spectrum brought about by the intense field were investigated as a function of the pressure in the range from 0.003 to 0.1 Torr. The power of the intense field was kept constant in the course of this (5 MW). It was found that at low pressure (Fig. 3Aa) the angular diagram of the scattered radiation on the red side of ω_2 is not at all pronounced and the angular divergence in this region increases with pressure.

At a pressure of 0.0038 Torr the widths of the lines of the resonance doublet ω_1, ω_2 are not great and are comparable with the values of the widths in the absence of the field (cf., Fig. 3). The dependence of the line widths on the pressure is shown in Fig. 4. The graph for $\Delta\omega_2$ refers to the outermost (along the height of the slit) part of the spectrum. In the central part the changes in $\Delta\omega_2$ within the same range of pressures are considerably smaller: 1.0 $cm^{-1} < \Delta\omega_2 < 1.5$ cm^{-1}. The following is noteworthy in the curves of Fig. 4. Firstly, the curves 3—5 showing the dependence of the line widths $\Delta\omega_1$, $\Delta\omega_2$, $\Delta\omega_3$ on p have a "break" which is situated at approximately the same pressure each time (0.01 Torr). Secondly, after the "break" the slope of the curves 4 and 5 is approximately the same as for the widths in the absence of the intense field (curves 1, 2). Thirdly, the maximum value of the broadening due to the field (i.e., of the difference of the curves of Fig. 4 with the field and without it) of the line ω_1 (~ 6 cm^{-1}) is twice as big as for the line ω_2 (~ 3 cm^{-1}). Finally, both these values are close to the width of the spectrum of the intense field ($\Delta\omega_S = 4$ cm^{-1}).

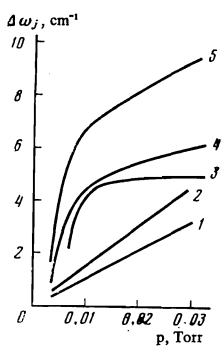

FIG. 4. Pressure dependence of the line widths $\Delta\omega_1$, $\Delta\omega_2$, $\Delta\omega_3$: The curves correspond to: 1) $\Delta\omega_1$ without the intense field, 2) $\Delta\omega_2$ without the intense field, 3) $\Delta\omega_3$ with the intense field, 4) $\Delta\omega_2$ with the intense field, 5) $\Delta\omega_1$ with the intense field.

The line ω_3 of the inverse Raman scattering appears at a pressure of 0.0068 Torr ($P_S = 5$ MW), and in Fig. 3 corresponding to the lower pressure of 0.0038 Torr it is absent. The width of this line also grows monotonically with the pressure (cf., Fig. 4) of potassium vapor from 2.0 cm^{-1} at 0.007 Torr to 5 cm^{-1} at p = 0.02 Torr. In contrast to the absorption lines ω_1 and ω_2, the line of inverse Raman scattering is practically no longer broadened with increasing pressure after the "break." If the pressure exceeds 0.1 Torr, the lines ω_2 and ω_3 overlap forming a single "absorption" band of width of the order of 20 cm^{-1}.

Spectrograms obtained in experiments with beams travelling in opposite directions are shown in Fig. 2B and Fig. 3B. These spectrograms correspond to the same pressures and to the same power of the intense field as in Fig. 2A and Fig. 3A. The majority of the effects of the intense field noted above, and in particular, the increased divergence of the DL radiation near ω_2 ("whiskers"), the field broadening of the lines of the resonance doublet ω_1 and ω_2, the change in the width of the line ω_2 along the height of the slit, the inverse Raman scattering line ω_3, has disappeared and was not observed over the whole range of pressures of potassium vapor 10^{-3} Torr < p < 0.1 Torr. Only a small field shift of the lines ω_1 and ω_2 (cf., Fig. 2Ba with Fig. 2Bb and Fig. 3Ba with Fig. 3Bb), approximately the same as in the case $\mathbf{k}_S \uparrow\uparrow \mathbf{k}_K$ at low pressure, is clearly observable.

4. DISCUSSION

We associate the observed effects in the frequency-angular distribution of the DL radiation in the presence of an intense field propagated in the same direction with the four-photon resonance parametric scattering (FRPS). Under our conditions (relatively great width of the spectrum of the intense field, $\Delta\omega_S = 4$ cm^{-1}; strongly resonant conditions) we assume the existence of FRPS of two kinds. In FRPS of the first kind two photons of the intense field are absorbed, and two photons of the weak (DL) field are emitted; in FRPS of the second kind one photon of the intense field and one photon of the weak field are absorbed, and also photons of both the intense and the weak field are emitted but at different frequencies. Processes of both types indicated above effectively proceed with conditions of both time and space phase synchronization being satisfied:

$$\omega_{i1}+\omega_{i2}=\omega_{f1}+\omega_{f2}, \tag{1a}$$

$$\mathbf{k}_{i1}+\mathbf{k}_{i2}=\mathbf{k}_{f1}+\mathbf{k}_{f2}. \tag{1b}$$

Under our conditions we have $|\omega_i - \omega_j|/\omega \lesssim 10^{-3}$, $|n_\omega - 1| \ll 1$, and formulas (1a), (1b) give the following relation between the angles $\theta_{i1,i2}$ and $\theta_{f1,f2}$ formed by the propagation vectors of the absorbed ($\mathbf{k}_{i1}$, $\mathbf{k}_{i2}$) and emitted ($\mathbf{k}_{f1}$, $\mathbf{k}_{f2}$) photons[2]:

$$\theta^2_{f1,f2}=\theta^2_{i1,i2}+\Delta n, \quad \Delta n=n_{f1}+n_{f2}-n_{i1}-n_{i2}. \tag{2}$$

We determine at what frequencies of the weak field is there an increased value of the nonlinear polarizability describing the FRPS. The spectrum of the intense field (SRS) is centered on a frequency which exceeds the frequency of the resonance transition by 12 cm^{-1}. Since $\omega_2 - \omega_1 = 58$ cm^{-1}, in the range of frequencies ω close

to ω_2 we can take into account one level $4P_{3/2}$ and neglect the influence of the level $4P_{1/2}$. In this approximation the following formula holds (cf., Appendix)

$$P^{nl}(\omega_{k1}) = \frac{4\pi}{c\hbar^3}\omega_{k1}|d_{02}|^2 N_{02}\left\{\frac{\Omega_{S1}+\Omega_{S2}}{\Omega_{k1}(\Omega_{S1}+\Omega_{S2}-\Omega_{k1})}\frac{E_{S1}E_{S2}E_{k3}^*}{\Omega_{S1}\Omega_{S2}} + \frac{\Omega_{S2}+\Omega_{k1}}{\Omega_{k1}(\Omega_{S2}-\Omega_{S1}+\Omega_{k1})}\frac{E_{S1}E_{S2}^*E_{k2}}{\Omega_{S1}\Omega_{S2}}\right\}, \quad (3)$$

$$\Omega_{ki}=\omega_{ki}-\omega_2, \quad i=1, 2, 3; \quad \Omega_{Sj}=\omega_{Sj}-\omega_2, \quad j=1, 2,$$

where d_{02} is the matrix element of the dipole moment, N_{02} is the difference in the populations of the levels $4S_{1/2}$ and $4P_{3/2}$; the indices Sj and ki denote quantities which refer to the intense and the weak fields.

The first term in (3) describes a process of the first type (the photons ω_{S1}, ω_{S2} disappear, the photons ω_{k1}, ω_{k3} are emitted), as a result of which the energy of the intense field is "converted" into radiation which coincides in frequencies with the DL radiation. Without restricting generality we can assume that $\omega_{k1} < \omega_{k3}$ or, taking (1a) into account, $\omega_{k1} < (\omega_{S1} + \omega_{S2})/2$. For simplicity we set $\theta_{S1,S2} = 0$. Then from (2) we obtain that $\omega_{k1} < \omega_2$, i.e., the photons of the weak field must be emitted on the long wavelength side of the line of resonance absorption ω_2. Further, it can be seen from (3) that the nonlinear polarizability increases as $\Omega_{k1} \to 0$ and $\Omega_{k3} \to \Omega_{S1} + \Omega_{S2}$. Consequently[1)], both the efficiency of the process and the angle at which photons of the weak field are emitted must increase as the frequency ω_{k1} moves from the red side towards the absorption line ω_2. We thus associate the "whiskers" lying in this region, i.e., the increased divergence and intensity of the DL radiation due to the intense SRS field (Fig. 2Aa), with the first term in expression (3) for $P^{nl}(\omega_{k1})$, i.e., with FRPS of the first kind.

From formula (2) it follows that at a fixed frequency ω_{k1} the square of the angle of emission of a photon of the weak field ($\theta^2_{k1,k3}$) increases linearly with pressure since $\Delta n \propto \alpha p$. This relationship was confirmed (with an accuracy of 25–30%). We recall that the same dependence on pressure has been established earlier for the position of the "whiskers" arising when the SRS radiation in nitrobenzene and in α-chloronaphthalene passes through potassium vapor[2,15].

The second term in expression (3) for the cubic polarizability corresponds to a process in which photons ω_{S1} and ω_{k2} disappear and photons ω_{k1} and ω_{S2} are emitted. Here, consequently, the total number of photons of the intense field is not altered, only a redistribution of them in the spectrum takes place. The same also applies to the weak field. Condition (1a) in this case has the form $\omega_{k1} - \omega_{k2} = \omega_{S1} - \omega_{S2}$, i.e., a redistribution of energy in the spectrum of the DL radiation due to FRPS of the second kind can occur in a region of the spectrum of width of the order of the width of the SRS spectrum (approximately 4 cm^{-1}).

We consider that field broadening of the lines ω_1, ω_2 is due specifically to FRPS of the second kind. Let us assume that those regions of the spectrum where in the FRPS process DL photons disappear, are related by the conditions of synchronism to the absorption bands. Then the intensity in this region of the spectrum will be diminished, and photons emitted in FRPS will be absorbed and can not manifest themselves in the spectrogram. Thus, FRPS of the second kind combined with the "usual" single-quantum absorption can lead to a broadening of the absorption lines by an amount of the order of magnitude of the width of the spectrum of the intense field. Using a picturesque expression, the resonance lines serve as a "sink" for photons arising in a FRPS process of the second kind.

We first consider the situation near the line ω_2, and for this we use formula (3). Let $\omega_{S2} > \omega_{S1}$; in this case condition (2) is satisfied for $\omega_{k1} < \omega_2 < \omega_{k2}$, i.e., a photon is absorbed from the blue side of ω_2, a photon ω_{k2} is emitted on the red side. From (3) it can be seen that the efficiency of the process increases as one approaches the absorption line ω_2 and the emitted photon will be absorbed in the transition $4S_{1/2} - 4P_{3/2}$. Now let $\omega_{S2} < \omega_{S1}$; then condition (2) is satisfied for $\omega_{k2} < \omega_{k1} < \omega_2$, i.e., the photon ω_{k1} emitted in FRPS lies closer to the absorption band ω_2 than the photon ω_{k2} which disappears in FRPS. Since the efficiency of the process increases as $\omega_{k1} \to \omega_2$, then in this case also the photons ω_{k1} which happen to be close to ω_2 will be absorbed.

We carry out a detailed analysis in the region of the spectrum adjacent to the absorption line ω_1. Here FRPS of the first kind will take place with a lower efficiency, since it includes a weak field at a relatively nonresonant frequency

$$\omega_{k2}-\omega_{S1}=\omega_{S2}-\omega_{k1}\approx\omega_S-\omega_1=70 \text{ cm}^{-1}.$$

Therefore we restrict ourselves to an analysis of the FRPS of the second kind which occurs with the same efficiency as in the neighborhood of the ω_2 line. The part of the nonlinear polarization which describes FRPS of the second kind near ω_1 has the form (cf. the Appendix)

$$P^{nl}(\omega_{k1}) = \frac{2\pi}{c\hbar^3}\omega_{k1}|d_{01}d_{02}|^2 E_{S1}E_{S2}^*E_{k2}\left\{\frac{\Omega_{k1}+\Omega_{S2}}{\Omega_{k1}(\Omega_{k1}+\Omega_{S2}-\Omega_{S1})}\times\frac{N_{02}}{\Omega_{S1}\Omega_{S2}}+\frac{N_{21}}{\Omega_{k1}\Omega_{k2}(\Omega_{k1}-\Omega_{S1})}\right\}, \quad (4)$$

$$\Omega_{Si}=\omega_{Si}-\omega_2, \quad \Omega_{kj}=\omega_{kj}-\omega_1,$$

where N_{21} is the difference in the populations of the $4P_{3/2}$ and $4P_{1/2}$ levels. The first term in (4) has the same structure as the second term in (3) and leads to an analogous effect—field broadening of the line ω_1. Thus, the fact of field broadening of the lines ω_1 and ω_2 is easily explained by the hypothesis concerning FRPS of the second kind.

The same hypothesis can also explain other characteristic features of field broadening (cf. Sec. 3). According to Fig. 4 the broadening of the line ω_1 is twice as great as that of the line ω_2. This fact is explained by the circumstance that the field broadening is determined not only by FRPS (the corresponding terms in (3) and (4) are approximately the same), but also by single-quantum absorption; and absorption in the presence of the SRS field is greater for the $4S_{1/2} - 4P_{1/2}$ transition, than for the $4S_{1/2} - 4P_{3/2}$ transition, since the intense field tends to equalize the populations of the levels in the second case to a greater degree.

From a discussion of formula (3) it follows that field broadening on the long wavelength side must be greater than on the short wavelength side. This conclusion is in complete agreement with the asymmetry of the line ω_1 (cf., Fig. 2Aa). The break in the curves of Fig. 4 is explained within the framework of our hypothesis in the following manner: in the low pressure region field

broadening manifests itself completely, i.e., to the widths of the lines ω_1, ω_2 a quantity of the order of $\Delta\omega_S$ (6 cm^{-1} for ω_1 and 3 cm^{-1} for ω_2) is added. For $p > 0.01$ Torr a further broadening occurs only as a result of an increase of the line width in the absence of the field. And for this reason also the slopes of the curves 1, 2, 4, 5 are approximately the same. The characteristic features of field broadening of the lines ω_1, ω_2 discussed above serve as weighty arguments in favor of our interpretation.

The second term in (4) also corresponds to FRPS of the second kind, but with resonance at the frequency of inverse Raman scattering ($\Omega_{k1} = \Omega_{S1} = \omega_{S1} - \omega_2$). The index of refraction in the neighborhood of ω_3 is determined by both transitions to approximately the same extent[3], its estimate is unreliable, and it is difficult to draw definite conclusions from (2). One might think, however, that this term produces a field broadening of the line ω_3 analogous to the one considered above: the FRPS redistributes the photons in the DL spectrum near ω_3, while the "sink" of photons provides for the inverse Raman scattering (instead of the single-quantum absorption in the case of lines ω_1 and ω_2).

An important argument confirming the essential role played by FRPS in the observed phenomena turns out to be provided by the results of our experiments with beams travelling in opposite directions: with oppositely directed k_K and k_S there are no manifestations of any kind of frequency-angular diffusion in the DL spectrum. This is as it should be from the point of view being developed here since in this case the condition of spatial phase synchronization is not fulfilled.

We summarize Secs. 3 and 4. When powerful resonance SRS radiation and a relatively weak "test" DL radiation of wide spectrum were propagated in the same direction in potassium vapor a number of changes were observed in the DL spectrum brought about by the intense field. In particular: an anomalous broadening was observed for the lines ω_1 and ω_2 of potassium and for the inverse Raman scattering line ω_3; the field broadening is different for the lines ω_1 and ω_2 and depends on the pressure; an increase in the divergence of the DL radiation occurs on the red side of ω_2 ("whiskers") and this phenomenon depends on the pressure of potassium vapor; all the effects enumerated above are absent when the SRS and the DL radiation are propagated in opposite directions. The whole set of phenomena including very detailed ones (for example, the difference in the field broadening of the components of the resonance doublet ω_1, ω_2), has received a unified explanation as a consequence of the four-photon resonance parametric scattering (FRPS).

In conclusion we discuss briefly the possibility of a different interpretation. We first recall that the cubic nonlinear polarizability can be written in the following form (in the absence of spatial dispersion, cf., for example,[16]):

$$P^{nl}(\omega, \mathbf{k}) = \int d\omega_1\, d\omega_2\, d\omega_3\, d\mathbf{k}_1\, d\mathbf{k}_2\, d\mathbf{k}_3\, \Theta(\omega, \omega_1, \omega_2) E(\omega_1, \mathbf{k}_1) E(\omega_2, \mathbf{k}_2) \times E^*(\omega_3, \mathbf{k}_3)\, \delta(\omega_1+\omega_2-\omega_3-\omega)\, \delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}). \quad (5)$$

If the spectrum of the field $E(\omega, \mathbf{k})$ is of sufficiently small width and the polarizability $\Theta(\omega, \omega_1, \omega_2)$ can be taken outside the integral sign, then after going over from a spectral description to a space-time description expression (5) reduces to the following

$$P^{nl}(t, \mathbf{r}) = \Theta(\omega) |E(t, \mathbf{r})|^2 E(t, \mathbf{r}). \quad (6)$$

With the aid of (6) one can describe self-focusing, AM-PM conversion[8], "degenerate" parametric scattering[17] and other effects of self-action. In the case of small width of the spectrum of the intense field and large width of the spectrum of the weak field one can carry out the inverse Fourier transformation only with respect to the intense field variables, and obtain

$$P^{nl}(\omega_K, \mathbf{r}) = \Theta_1(\omega_K, \omega_S) E_S^2(\mathbf{r}) E_K^*(2\omega_S - \omega_K, \mathbf{r}) + \Theta_2(\omega_K, \omega_S) |E_S(\mathbf{r})|^2 E_K(\omega_K, \mathbf{r}), \quad (7)$$

where E_S, E_k are respectively the amplitudes of the intense and the weak fields. The first term in (7) describes four-photon scattering of the first kind, and the second term describes the effect of the intense field on the index of refraction for frequencies of the weak field (ω_k). Formula (7) serves as the basis for the interpretation of experimental facts under nonresonance conditions.

The phenomena investigated in the present work occur at distances of several cm^{-1} from the resonance absorption lines ω_1, ω_2 and the Raman scattering line ω_3. From formulas (3), (4) it can be seen that within the limits of the width of the spectrum of the intense field ($\Delta\omega_S \sim 4$ cm^{-1}) the polarizability Θ can vary by very-large amounts. Therefore under our conditions the transition from (5) to (7) is impossible, and the phenomena must be classified in terms of the language of FRPS: the intergrand in (5) states that photons $(\omega_1, \mathbf{k}_1)$ and $(\omega_2, \mathbf{k}_2)$ disappear and photons $(\omega, \mathbf{k})$ and $(\omega_3, \mathbf{k}_3)$ are emitted when the condition

$$\omega + \omega_3 = \omega_1 + \omega_2, \quad \mathbf{k} + \mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2,$$

analogous to (1) is satisfied. In this case the second term in (7) corresponds to FRPS of the second kind.

Particularly clearly the role played by resonance can be seen on the example of field broadening of the lines ω_1, ω_2, ω_3. In accordance with our hypothesis field broadening is brought about by FRPS of the second kind, with the emitted photon falling into the single-quantum absorption band (cf., discussion of formulas (3) and (4)) or into the band of inverse Raman scattering. The conditions for field broadening of the short wavelength side of the absorption band ω_1, ω_2 are of an "ultrasonant" nature: here a photon disappearing in the FRPS process is situated on one side of the frequency of the atomic transition, while the photon emitted in FRPS is situated on the other side.

Thus, both the analysis of specific experimental data and also general considerations support the hypothesis of the origin of the phenomena observed by us as a result of four-photon resonance parametric scattering (FRPS).

The FRPS investigated in the present work can have definite significance in many situations. We think, for example, that field broadening of infrared emission lines originating in potassium vapor under closely similar conditions[18,19] can be brought about by FRPS of the second kind. Further, the asymmetry of field broadening of the line ω_1 indicates that FRPS can mask or significantly distort the field (the so-called Stark) line shift. Special measurements of field shifts of six lines in the neighborhood of the resonance doublet of potassium[20,15] (one-, two-, and three-photon lines) have shown real deviations from the predictions of theory. Some of them are apparently associated with FRPS of the first kind (for ω_3) and of the second kind (for ω_1).

APPENDIX

We consider a two-level system (0, 2), which is acted upon by the electromagnetic field of the (four-frequency) form

$$E(t,\mathbf{r})=E_{S1}\exp\{i(\mathbf{k}_{S1}\mathbf{r}-\omega_{S1}t)\}+E_{S2}\exp\{i(\mathbf{k}_{S2}\mathbf{r}-\omega_{S2}t)\}+E_{K1}\exp\{i(\mathbf{k}_{K1}\mathbf{r}-\omega_{K1}t)\}+E_{K2}\exp\{i(\mathbf{k}_{K2}\mathbf{r}-\omega_{K2}t)\}. \quad (A.1)$$

We shall be interested in the nonlinear cubic polarization on the assumption that E_{S1}, E_{S2} represent intense and E_{k1}, E_{k2} represent weak fields:

$$|E_{Ki}|\ll|E_{Si}|. \quad (A.2)$$

Therefore we retain terms linear in E_{ki} and quadratic in E_{Si}. When the conditions

$$\omega_{S1}+\omega_{S2}=\omega_{K1}+\omega_{K2},\quad \mathbf{k}_{S1}+\mathbf{k}_{S2}=\mathbf{k}_{K1}+\mathbf{k}_{K2} \quad (A.3)$$

are satisfied the nonlinear polarization turns out to be equal to

$$P_1^{nl}(\omega_{K1})=\frac{4\pi}{c\hbar^3}\omega_{K1}|d_{02}|^4N_{02}\frac{\Omega_{S1}+\Omega_{S2}}{\Omega_{S1}\Omega_{S2}\Omega_{K1}\Omega_{K2}}E_{S1}E_{S2}E_{K2}^*, \quad (A.4)$$

$$\Omega_{Si}=\omega_{Sj}-\omega_2,\quad \Omega_{Ki}=\omega_{Kj}-\omega_2.$$

The nonlinear polarization for FRPS of the second kind is obtained from (A.4) with the aid of the replacements

$$E_{S2}\to E_{K3},\quad E_{K2}^*\to E_{S2}^*.$$

Thus, when the conditions

$$\omega_{S1}+\omega_{K3}=\omega_{S2}+\omega_{K1},\quad \mathbf{k}_{S1}+\mathbf{k}_{K3}=\mathbf{k}_{S2}+\mathbf{k}_{K1}, \quad (A.5)$$

analogous to (A.3) are satisfied we obtain

$$P_2^{nl}(\omega_{K1})=\frac{4\pi}{c\hbar^3}\omega_{K1}|d_{02}|^4N_{02}\frac{\Omega_{S1}+\Omega_{K3}}{\Omega_{S1}\Omega_{S2}\Omega_{K1}\Omega_{K3}}E_{S1}E_{S2}^*E_{K3}. \quad (A.6)$$

We now consider the three-level system (0, 1, 2) of energies (E_0, E_1, E_2) in which dipole transitions 0-1 and 0-2 are allowed and upon which the field (A.1) is acting. If

$$\omega_2\gg|\omega_2-\omega_1|\gg|\omega_2-\omega_S|,$$

then polarization at frequencies ω_k close to ω_2 ($|\omega_k-\omega_2|\ll|\omega_2-\omega_1|$), can be calculated without taking into account the 0-1 transition, i.e., in accordance with formulas (A.3)—(A.6) for a two-level system. And near the frequency of the other 0-1 transition ($|\omega_k-\omega_1|\ll|\omega_2-\omega_1|$) due to the coupling of the transitions 0-2, 0-1 through the common level 0 a nonlinear polarization of parametric origin arises:

$$P^{nl}(\omega_{K1})=\frac{2\pi}{c\hbar^3}\omega_{K1}|d_{01}d_{02}|^2\frac{E_{S1}E_{S2}^*E_{K2}}{\Omega_{K1}\Omega_{K2}}\times\left\{\frac{\Omega_{S2}+\Omega_{K1}}{\Omega_{S1}\Omega_{S2}}N_{01}+\frac{N_{21}}{\Omega_{K1}-\Omega_{S1}}\right\}; \quad (A.7)$$

$$\Omega_{Ki}=\omega_{Ki}-\omega_1,\quad \Omega_{Si}=\omega_{Si}-\omega_2.$$

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