Field Splitting of the Potassium Violet Lines

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The hypothesis of the four-photon parametric nature of the violet radiation from potassium atoms (λ 4044/47 Å, $5P_{3/2,1/2}-4S_{1/2}$ transitions) induced by irradiation with a strong ruby laser field and its Stokes stimulated Raman scattering component in nitrobenzene is experimentally confirmed. Self-focusing in potassium vapor of ruby laser stimulated Raman scattering radiation is observed. A theoretical analysis is presented of the general laws of term splitting in multilevel systems located in several strong monochromatic fields. The structure of the violet radiation is interpreted as being the result of field splitting of terms.

1. INTRODUCTION

IN[1] we reported observation of a structure in the violet radiation (λ = 4044/47 Å, transitions $5P_{3/2,1/2}-4S_{1/2}$, Fig. 1) of potassium vapor simultaneously irradiated by a giant ruby-laser pulse (ω_R) and the Stokes SRS of its radiation in nitrobenzene (ω_S). The ruby-laser radiation is in resonance with the transition $4P_{3/2}-6S_{1/2}$, and the SRS radiation is at resonance with the transition $4S_{1/2}-4P_{3/2}$. The violet radiation was interpreted as a four-photon parametric process in which the photons ω_R and ω_S are absorbed, and an infrared photon ω_i (transition $6S_{1/2}-5P_{3/2}$) and a violet photon ω_V are emitted. The structure of the violet radiation, which takes the form of dips in the emission line, was interpreted as components of the absorption line arising as a result of the field splitting of the $5P_{3/2}$ term. We attributed this splitting to the interaction of the potassium atoms with three powerful monochromatic waves: the ruby-laser radiation field, the infrared-radiation field of the stimulated electronic Raman scattering (SERS) by the potassium atoms (transitions $4P_{3/2}-6S_{1/2}-5P_{3/2}$) and the infrared radiation field at resonance with the transition $6S_{1/2}-5P_{3/2}$.

Our investigations[2] of the potassium-vapor infrared emission have made it possible to refine and supplement the hypothetical three-field model. All the lines of the cascade $6S_{1/2}-4S_{1/2}$ which are compatible with the selection rules (see the table) were observed. The most intense among them are the lines corresponding to the transitions $5P_{3/2,1/2}-3D_{3/2}$ and $5P_{3/2}-5S_{1/2}$. The integral power of the strongest among them is ~ 10 kW. It should be noted that the SERS line was observed only at sufficiently large vapor pressures p and strong-field powers P_R ($pP_R \gtrsim 1$ Torr-MW/cm^2). When the SERS line appears, the intensities of the radiation fields at the transitions $3D_{5/2,3/2}-4P_{3/2}$, $5S_{1/2}-4P_{3/2}$ decrease sharply, the violet-radiation doublet changes into a multicomponent structure, and "short-wave" lines appear[1].

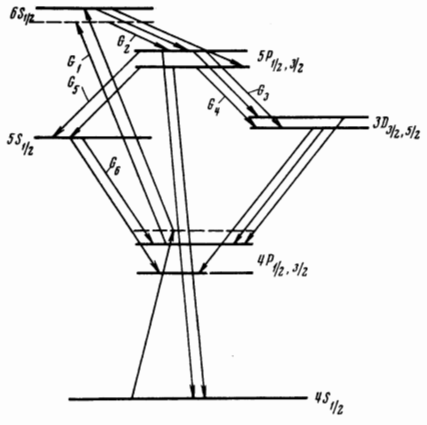

FIG. 1. Scheme of potassium terms.

The line at resonance with the transition $6S_{1/2}-5P_{3/2}$ has a large spectral width (up to 3 cm^{-1}) and therefore cannot be connected with the observed structure of the violet radiation.

The present investigation had several purposes. First, to verify the hypothesis of the four-photon nature of the violet radiation, for which purpose an "opposing" beam experiment was performed (Sec. 2). Second, to observe in potassium vapor the self-focusing of the Stokes SRS of ruby-laser radiation in nitrobenzene; this phenomenon, observed by Bonch-Bruevich et al.[3] and by Grischowsky[4] in potassium vapor, can strongly influence the splitting of the atomic terms (Sec. 2). Since, furthermore, the experiments performed in[2] revealed a large number of strong quasimonochromatic

Transitions	Line frequency, cm^{-1}	Line width, cm^{-1}	Transitions	Line frequency, cm^{-1}	Line width, cm^{-1}
$6S_{1/2}-5P_{3/2}$	2730.4	1.5–3.0	$5P_{1/2}-3D_{3/2}$	3167.0	0.5
$6S_{1/2}-5P_{1/2}$	2749.2	0.8	$3D_{3/2}-4P_{3/2}$	8491.5	1.2
$5P_{3/2}-5S_{1/2}$	3693.2	1.2	$3D_{3/2}-4P_{3/2}$		
$5P_{1/2}-5S_{1/2}$	3674.4	1.0	$3D_{3/2}-4P_{1/2}$	8549.2	1.2
$5P_{3/2}-3D_{5/2}$	3183.45	0.5	$5S_{1/2}-4P_{3/2}$	7984.1	1.3
$5P_{3/2}-3D_{3/2}$	3185.8	0.5	$5S_{1/2}-4P_{1/2}$	8041.8	1.5
			$4P_{3/2}-5P_{3/2}$ (SERS)	2722.3	0.5

radiation fields at coupled transitions, it was necessary to analyze the general laws governing multilevel systems in several strong monochromatic fields (Sec. 3). Finally, in Sec. 4 we interpret the structure of the violet lines.

2. "OPPOSING BEAM" EXPERIMENT AND OBSERVATION OF SELF-FOCUSING

Four-photon parametric processes are effective if the following conditions are satisfied (see, for example,[5]):

$$\omega_s + \omega_R = \omega_i + \omega_v, \quad \mathbf{k}_s + \mathbf{k}_R = \mathbf{k}_i + \mathbf{k}_v,$$

where $\mathbf{k}$ are the wave vectors of the beams taking part in the process. If $\mathbf{k}_S$ and $\mathbf{k}_R$ are parallel ($\mathbf{k}_S \uparrow\uparrow \mathbf{k}_R$), then both $\mathbf{k}_i$ and $\mathbf{k}_v$ should be parallel to them, as was observed by us in[1,2], namely, both the violet and the IR emissions propagate only in the direction of $\mathbf{k}_S$ and $\mathbf{k}_R$ when $\mathbf{k}_S \uparrow\uparrow \mathbf{k}_R$. On the other hand, in the case of antiparallel $\mathbf{k}_S$ and $\mathbf{k}_R$ ($\mathbf{k}_S \uparrow\downarrow \mathbf{k}_R$), one should expect a sharp decrease in the effectiveness of the parametric process: since $|\mathbf{k}_S|$ and $|\mathbf{k}_R|$ are close, and $|\mathbf{k}_i|$ and $|\mathbf{k}_v|$ differ strongly ($\omega_v = 24\,720.2$ cm^{-1}, $\omega_i = 2730.45$ cm^{-1}), the phase synchronism condition is not satisfied for $\mathbf{k}_S \uparrow\downarrow \mathbf{k}_R$, and the length of the coherent interaction has the same magnitude as the wavelength.

The experimental setup for the "opposing beam" experiments is shown in Fig. 2. The beam from a 70-MW laser is split by prisms into two parts of equal cross section, which are guided with the aid of prisms 3 and 11 and dielectric mirrors 6 and 8 from opposite directions into a cell 7 filled with potassium vapor. The cell 7 was either 20 or 60 cm long. The reflectances of mirrors 6 and 8 were close to unity in the red region of the spectrum and 0.2–0.3 in the violet region. A cell with nitrobenzene, 20 cm long, could be placed in the path of the first (1, 11, 8) or second (2, 3, 6) beam, in position 4 or 10. The filter 5 (FS-6) separated the Stokes SRS. The slit of the DFS-8 spectrograph was in the focal plane of the lens 9. The registration was photographic, photoelectric, or visual. The experimental setup was symmetrical. The beam path 1, 11, 8, 7 was equivalent to the path 2, 3, 6, 7. This was attained by using identical prisms (1, 3, 11) and mirrors (6, 8) and placing them at equal distances relative to the unsplit laser beam. The setup made it possible to maintain the same laser and SRS radiation density in the cell with potassium when the directions of $\mathbf{k}_R$ and $\mathbf{k}_S$ were varied.

The experiment was performed in the following sequence. Filter 5 was first removed and the laser-beam radiation ($\mathbf{k}_R$) and the SRS radiation ($\mathbf{k}_S$) were parallel ($\mathbf{k}_R \uparrow\uparrow \mathbf{k}_S$) and followed the path 1, 11, 10, 8. The other path (2, 3, 5, 7) was blocked by an opaque screen. In this case the intense violet glow of the potassium vapor was registered in a direction coinciding with $\mathbf{k}_S$ and $\mathbf{k}_R$, and was not observed at all in the opposite direction. Analogously, for $\mathbf{k}_R \uparrow\uparrow \mathbf{k}_S$ and the radiation propagating along the other path (2, 3, 4, 6), the violet radiation is also observed only in the forward direction coinciding with that of $\mathbf{k}_S$ and $\mathbf{k}_R$.

If the transparent screen is now removed and the filter 5 introduced, then beams with $\mathbf{k}_S \uparrow\uparrow \mathbf{k}_R$ propagate

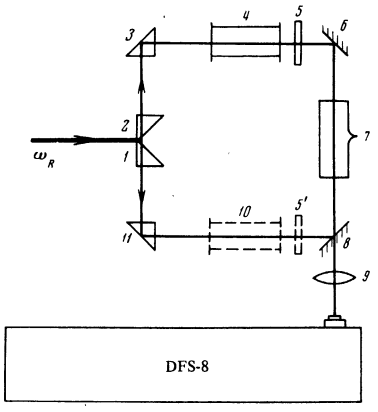

FIG. 2. Diagram of experimental setup.

in the cell 7. In this case no violet radiation was registered. The observation was made in the directions of $\mathbf{k}_R$ and $\mathbf{k}_S$ simultaneously, and there was no radiation in either direction.

The experiments were performed with a threefold excess of the pump over the threshold of excitation of the violet lines (for $\mathbf{k}_S \uparrow\uparrow \mathbf{k}_R$). The recording apparatus had a 50-fold gain margin (relative to the observation threshold). To perform the opposing-beam experiment, the laser beam was guided along the path 1, 11, 8 and its Stokes SRS in the nitrobenzene along the path 2, 3, 4, 5, 6. Simultaneously with the violet radiation, we photographed the Stokes SRS spectrum (ω_S) with the aid of the DFS-8 spectrograph. All the spectrograms of this radiation revealed clearly the two-photon absorption band, and consequently the atoms went over from the state $4S_{1/2}$ into $6S_{1/2}$. A fact confirming the population of the $6S_{1/2}$ level was the observation of IR emission lines of the potassium vapor at the transitions $6S_{1/2}$–$5P_{3/2,1/2}$ etc.[2] also in the case of the opposing beams. Variations of the experimental conditions (change of the vapor pressure, of the power, and of the radiation frequency of the strong fields ω_R and ω_S) has shown that the violet channel is always completely "blocked" when $\mathbf{k}_R \uparrow\downarrow \mathbf{k}_S$, and the IR radiation is always observed, but with an intensity lower than in the case when $\mathbf{k}_R \uparrow\uparrow \mathbf{k}_S$.

The absence of violet radiation when $\mathbf{k}_S \uparrow\downarrow \mathbf{k}_R$ may be connected with the fact that there is no inverted population between the levels $5P_{3/2}$ and $4S_{1/2}$ ($4S_{1/2}$ is the ground level), and consequently there are no reasons for superradiance. On the other hand, the parametric process, as already noted, is not very effective. At the same time, there is population inversion between the levels $6S_{1/2}$–5P–5S–(3D), and we observe the superradiance effect.

Thus, the experiments have shown convincingly that the cause of the appearance of violet lines is four-photon parametric scattering, in which photons of the laser emission (ω_R) and of the SRS (ω_S) are absorbed, and photons of infrared (transition $6S_{1/2}$–$5P_{3/2}$) and violet (transition $5P_{3/2}$–$4S_{1/2}$) frequencies are emitted.

The self-focusing of the ω_S radiation in a resonant medium (potassium vapor) was observed in[3,4]. This phenomenon was also observed in our experiments, but under somewhat different conditions. Taking into account

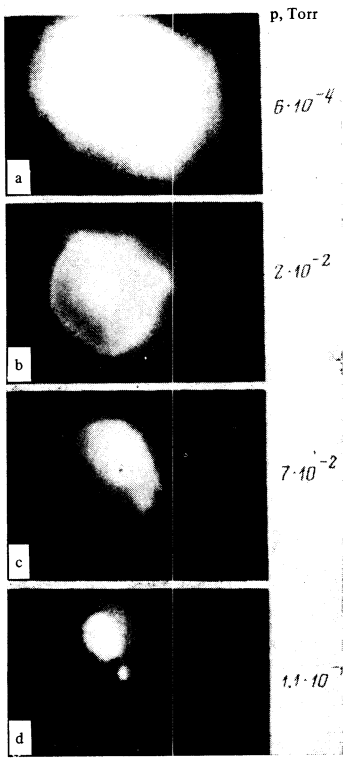

FIG. 3. Change in the distribution of the SRS and violet-radiation intensity at the output window of the cell, as a function of the potassium vapor pressure.

the fact that the ω_S radiation is closely connected with excitation of all the emission lines of the potassium vapor observed by us in[2], we present here certain results of an investigation of this phenomenon.

We were interested in the change of the intensities of ω_S and ω_V over the cross section after passing through the cell with potassium. The experiments were performed with the setup described in[1]. The cell with potassium was 60 cm long. We photographed simultaneously the input and output windows of the cell with potassium on a photographic film having a spectral sensitivity range 0.4—0.75 μ. The experimental results are shown in Fig. 3. The minimum cross sections of the distribution of the ω_S intensity (Fig. 3d) were observed at potassium vapor pressures $p = 1.1 \times 10^{-1}$ Torr.

Measurements of the spot areas have shown that the density of the radiation power in the spots (W/cm^2) increases by 40—50 times. It was established that the intensity distribution over the cross section of the violet radiation (ω_V) coincides with the distribution for ω_S. On this basis it can be assumed that the spatial distribution of the IR line intensity coincides with the distribution of the SRS intensity.

3. SPLITTING OF TERMS OF THE MULTILEVEL SYSTEM IN SEVERAL MONOCHROMATIC FIELDS

Before we turn to concrete models that determine the splitting of the terms of potassium, let us stop to discuss a number of laws governing the term splitting of multilevel systems in several resonant fields acting on adjacent transitions. We consider a system of n

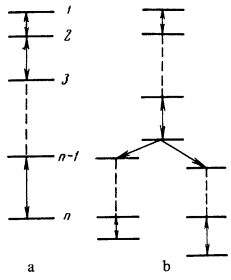

FIG. 4. Level scheme of n-level atom interacting with a polymonochromatic field.

levels (Fig. 4a) in which each transition between levels is acted upon by a strong monochromatic field whose deviation from the transition frequency is $\Omega_j = \omega_j - \omega_{0j}$, where $|\Omega_j| \ll \omega_{0j}$ (ω_j is the field frequency and ω_{0j} is the transition frequency). The wave function of such a system will be sought in the usual form

$$\Psi(\mathbf{r}, t) = \sum_{j=1}^{n} a_j(t)\psi_j(\mathbf{r})\exp\left(\frac{E_j t}{i\hbar}\right). \tag{1}$$

Making the change of variables

$$a_j(t) = b_j(t)\exp\left\{it\sum_{m=1}^{j-1}\Omega_m\right\},$$

we obtain for the coefficients $b_j(t)$ a system of ordinary differential equations with constant coefficients. We write it in matrix form

$$i\dot{\hat{b}} = \hat{B}\hat{b} \tag{2}$$

$$\hat{b}(t) = \begin{pmatrix} b_1(t) \\ b_2(t) \\ \cdots \\ b_n(t) \end{pmatrix}, \quad \hat{B} = \begin{pmatrix} 0 & G_{12} & & & \\ G_{21} & \Omega_1 & G_{23} & & \\ & G_{32} & \Omega_1+\Omega_2 & G_{34} & \\ & & \cdots & & \\ & & & G_{n,n-1} & \Sigma \end{pmatrix}$$

where

$$G_{ij} = \frac{1}{2\hbar}E_j d_{ij}, \quad \Sigma = \sum_{j=1}^{n-1}\Omega_j.$$

We seek the solution in the form $\hat{b}(t) = e^{i\epsilon t}\hat{b}_0$, where $\hat{b}_0$ is a constant vector. Substituting the solution in such a form in (2), we obtain an algebraic system of equations

$$(\hat{B} + \varepsilon\hat{E})\hat{b}_0 = 0. \tag{3}$$

The roots of the determinant of the system (3) determine (together with $\Sigma\Omega_j$) the differences between the new natural frequencies of the system situated in the field and the frequencies of the unperturbed term, and specify the position of the split components of the term. The number of components is obviously equal in the general case to the order of the determinant. In the case of strict resonance (all $\Omega_j = 0$), the number of components may turn out to be $n-1$, i.e., the splitting will be incomplete. Let us consider the case of strict resonance.

The determinant of the system (3) is given by

$$D_n(\varepsilon) = \begin{vmatrix} \varepsilon & G_{12} & & & \\ G_{21} & \varepsilon & G_{23} & & \\ & G_{32} & \varepsilon & G_{34} & \\ & & \cdots & & \\ & & & G_{n-1,n-2} & \varepsilon & G_{n-1,n} \\ & & & & G_{n,n-1} & \varepsilon \end{vmatrix}.$$

Expanding it in terms of the last line, we obtain the recurrence relation

$$D_n(\varepsilon) = \varepsilon D_{n-1}(\varepsilon) - |G_{n-1,\,n}|^2 D_{n-2}(\varepsilon). \quad (4)$$

Since $D_0(\epsilon) = 1$ and $D_1(\epsilon) = \epsilon$, we have

$$D_2(\varepsilon) = \varepsilon^2 - |G_{12}|^2, \quad D_3(\varepsilon) = \varepsilon^3 - [|G_{12}|^2 + |G_{23}|^2]\varepsilon \quad (4')$$

etc. We can see from (4) and (4′) that for odd n the determinant $D_n(\epsilon)$ has one zero root; it is easy to show that it corresponds to the zero solution of the system (3), and consequently the number of components will be $n-1$, i.e., the splitting is incomplete. For even n, there are no zero roots and the splitting is complete.

Let us examine an n-level branching system (Fig. 4b). Assume that each of the transitions is acted upon by a strictly resonant field. An analysis of the roots of the determinant of such a system, based on recurrence relations similar to (4), gives a splitting picture which is best written down graphically (Fig. 5). A line denotes a branch with indicated parity of the number of levels in it, and the central level (which is common to all branches) is not included when the levels are counted. The number of splitting components of each level is marked at the bottom.

The results are perfectly valid for branching systems in which the frequency deviations at two adjacent transitions in one of the branches are not equal to zero, but are equal and opposite in magnitude ($\Omega_j \neq 0$, $\Omega_j + \Omega_{j-1} = 0$). In the case of arbitrary deviations, the splitting will be complete.

In the case of an n-level system situated in strictly resonant fields and having one central level and $n-1$ branches, each of which contains one level, the splitting consists of two components and is given by the expression

$$\varepsilon_{1,2} = \pm\left[\sum_{j=2} |G_{1j}|^2\right]^{1/2}. \quad (5)$$

It is seen from (5) that the addition of new branches containing one level each to the central level does not change the doublet structure of the splitting, but only increases the magnitude of the splitting.

We present formulas for the splitting component of the terms of several systems (Fig. 6). These formulas will be needed for a subsequent analysis of the splitting of the terms in potassium. We note beforehand that the systems shown in Figs. 6a and b are formally equivalent to linear systems (Fig. 4), and therefore the splitting in them coincides with the splitting in linear systems containing the same number of levels. In the system shown in Fig. 6a, incomplete splitting into two components is realized. The positions of the components relative to the unperturbed term are given by the expression

$$\varepsilon_{1,2} = \pm\sqrt{|G_{12}|^2 + |G_{23}|^2}. \quad (6)$$

In the system of Fig. 6b, the splitting is also incomplete, and the positions of the four components are given by the expression

$$\varepsilon_{1,2,3,4} = \pm\left\{\frac{1}{2}\sum_{i=1}^{4}|G_{ii+1}|^2 \pm \left(\frac{1}{4}\left[\sum_{i=1}^{4}|G_{ii+1}|^2\right]^2 - |G_{12}G_{34}|^2 - |G_{12}G_{45}|^2 - |G_{23}G_{45}|^2\right)^{1/2}\right\}^{1/2}. \quad (7)$$

If all the $|G_{ii+1}|$ are equal ($|G_{ii+1}| = |G|$), we have

$$\varepsilon_{1,2} = \pm\sqrt{3}|G|, \quad \varepsilon_{3,4} = \pm|G|. \quad (7')$$

Expression (7′) can be regarded as the effective splitting of the components of the doublet (6) into pairs of sublevels with allowance for the fields G_{34} and G_{45}, and in this case the distance between the sublevels in the pair is equal to

$$|\varepsilon_1 - \varepsilon_3| = |\varepsilon_2 - \varepsilon_4| = (\sqrt{3} - 1)|G|,$$

and the distance between the pairs is

$$^1/_2[|\varepsilon_1 + \varepsilon_3| + |\varepsilon_2 + \varepsilon_4|] = (\sqrt{3} + 1)|G|.$$

The distance between the outermost components of the splitting is $2\sqrt{3}|G|$. This is larger than in the system in Fig. 6a, where, as follows from (6), this distance is equal to $2\sqrt{2}|G|$ under the condition $|G_{12}| = |G_{23}| = |G|$.

Under definite conditions (see below), the structure of the splitting (7′) can be resolved as a doublet. Such a specific picture of the splitting is not universal and is a consequence of the assumptions made concerning the values of the matrix elements G_{ii+1}. Thus, under the condition $|G_{12}| \gg |G_{ii+1}|$ for all $i \neq 1$ it follows from (7) that the splitting will consist of two close components split by an amount $\sim 2|G_{ii+1}|$, $i \neq 1$, and two remote ones, separated by a distance $2|G_{12}|$. The system shown in Fig. 6c pertains to type c of the branching systems of Fig. 5; the splitting in it is incomplete and it consists of four components. The position of the components of the splitting of level 3, under the condition $|G_{34}| = |G_{35}| = |G|$, $|G_{ii+1}| \ll |\Omega|$, is given for all i by the formulas

$$\varepsilon_{1,2} \cong \pm\sqrt{2}|G|, \quad \varepsilon_3 = (|G_{12}|^2 + |G_{23}|^2)/|\Omega|, \quad \varepsilon_4 = -|\Omega|. \quad (8)$$

Let us list briefly the main results of the present section. In many-level systems in which strictly resonant fields act on several transitions, incomplete splitting may be realized. In the case of nonzero frequency deviations Ω_j, the splitting may turn out to be incomplete only for certain particular values of Ω_j, and at arbitrary

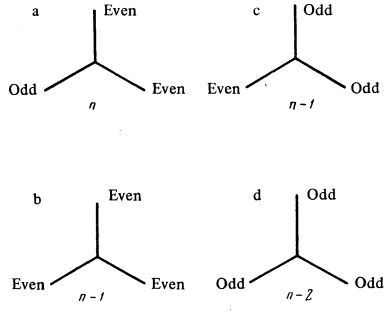

FIG. 5. Different types of branching systems.

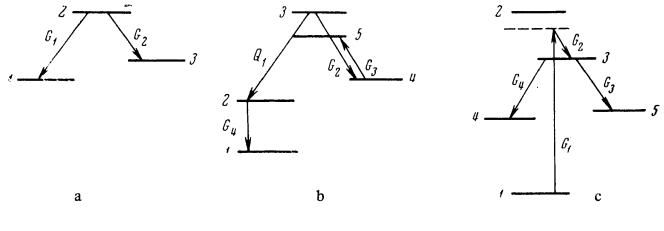

FIG. 6. Certain particular cases of the scheme of Fig. 4a.

deviations the splitting is complete. In the case of incomplete splitting in many-level systems, allowance for different numbers of levels coupled by strong fields can yield the same number of components, and only the magnitude of the splitting varies. It is therefore not always possible to associate the observed radiation structure uniquely with some concrete splitting model. Allowance for a large number of fields can lead not only to an increase in the number of components but also to an increase of the distance between them (see (5), (6), (7), (7′)).

4. DISCUSSION AND INTERPRETATION OF EXPERIMENTAL RESULTS

The dependence on the potassium vapor pressure and on the laser radiation power for the number of splitting components of the violet lines indicates that there is no universal model of field splitting. When the potassium vapor pressure and the radiation power are below the threshold for the appearance of the SERS ($pP_R < 1$ Torr-MW/cm^2), a doublet structure of the ultraviolet radiation absorption is observed[1]. Such a structure can be associated with the presence of strong monochromatic fields G_3 and G_5 (Fig. 1). The splitting components are given by (6). When the fields G_6 and G_4 are taken into account, we obtain the system shown in Fig. 6b. The splitting components are then given by expression (7), and in the approximation $|G_{ii+1}| = |G|$ they are given by formulas (7′). It is seen from (7′) that at a dip width $2|G| > \Delta\omega > (\sqrt{3} - 1)|G|$ this structure can be resolved as a doublet.

At pressures and powers $pP_R > 1$ Torr-MW/cm^2, an SESR line appears, and the violet radiation acquires a multicomponent structure. Three closely located dips and one dip at a distance of 8 cm^{-1} on the short-wave side appear. Such a structure can be associated with the action of the fields (Fig. 1) G_1, G_2, G_3, and G_5; we obtain the system shown in Fig. 6c. The splitting components are given by expressions (8), where $|\Omega| = 8$ cm^{-1}.

The experimentally observed infrared-field powers amount to ~10 kW. Here, for example, for the transition $5P_{3/2}$—$5S_{1/2}$ (oscillator strength f = 0.5, wavelength $\lambda = 2.7\ \mu$), the characteristic splitting parameter for a beam cross section of 1 cm^2 is 0.3 cm^{-1}, which is smaller by one order of magnitude than the distance between the structure components of the 4044-Å line.

In this estimate we assumed the cross section of the infrared radiation beam to be equal to the initial cross section of the Stokes SRS radiation. But since it becomes self-focused on passing through the potassium vapor, and the processes that lead to the occurrence of infrared lines (two-photon absorption, SERS, six-photon processes, see[2] for details) occur most effectively in regions of high intensity of the Stokes SRS, the registered infrared-radiation power actually comes from cross sections much smaller than the initial cross section of the Stokes SRS beam. This increases the estimated infrared field intensity E (see Sec. 2) by almost one order of magnitude. In addition, as shown in Sec. 3, the action of several powerful fields on coupled transitions leads to an increase of the splitting; thus, the characteristic parameter |G| in formulas (6) and (7′) for the magnitude of the splitting is multiplied by $\sqrt{2}$ and $\sqrt{3}$, respectively.

The performed analysis of the λ 4044/47 Å violet lines shows that this structure, just as in[1], should be attributed to field splitting of the $5P_{3/2}$ term. At the same time, the concrete three-field model advanced in[1] as a hypothesis apparently does not correspond to reality, by virtue of the large width of the line of one of the fields. However, the sharp IR lines observed in[2] (see the table and Fig. 1 of the present paper) explain the observed splitting qualitatively. A quantitative comparison of the experimental and theoretical data on the splitting is made difficult by the self-focusing of the SRS radiation in the potassium vapor.

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