Stimulated processes in potassium vapor in the presence of a buffer gas

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Stimulated resonant emission ($4P_{1/2}\rightarrow 4S_{1/2}$) transition and stimulated electronic Raman scattering ($4P_{1/2}\rightarrow 4P_{3/2}$) transition in potassium vapor in the presence of an He gas buffer are investigated experimentally and theoretically. The dependences of the emission energies on the intensity and frequency of the exciting radiation and on the buffer-gas and potassium-vapor pressures are obtained in experiment. It is shown that these processes can be satisfactorily described using a three-level laser-system model.

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Collisional relaxations play an important role in the spectral redistribution of quasiresonant radiation that interacts with gaseous media. Investigations of such processes are important both for fundamental and for applied problems in physics, particularly for the development of new coherent-radiation sources.[1–3]

Stimulated resonant emission connected with transitions between excited levels in atomic metal vapors in the presence of collisions was observed in Refs. 4 and 5. In the processes investigated, the final levels for the stimulated transitions were practically unpopulated.

In Refs. 6 and 7 was first reported the production of intense stimulated resonant emission (IRE) on the transition $4P_{1/2}\rightarrow 4S_{1/2}$ (the D_1 line) and stimulated electronic Raman scattering (SERS) connected with the transition $4P_{1/2}\rightarrow 4P_{3/2}$ (Stokes component) in a mixture of potassium vapor with a buffer gas, excited by radiation that is quasiresonant with the $4S_{1/2}\rightarrow 4P_{3/2}$ transition (D_2 line). Similar observation of IRE in sodium vapor in the presence of helium was reported in Ref. 8. The results of these studies shows that the collisions of potassium atoms on the $4P_{3/2}$ level with the buffer-gas atoms can lead to effective population and inversion of the $4P_{1/2}$ level relative to the ground level. This gives grounds for considering the set of potassium levels $4S_{1/2}$, $4P_{1/2}$, and $4P_{3/2}$ in the presence of collisions as a three-level laser system.[9]

We present here the results of theoretical and experimental investigations of the IRE and SERS processes in potassium vapor in the presence of a buffer gas. In Sec. I we analyze the expressions obtained from the balance equations for the level population differences, and in Sec. II we describe the experimental setup. The results of the experimental investigations and their discussions are contained in Sec. III.

I. We consider three-level atoms with energy levels $E_1 < E_2 < E_3$ in the field of laser radiation that is quasiresonant with the $1\rightarrow 3$ transition, in the presence of collisions with buffer-gas atoms. Let the transitions $1\rightarrow 3$ and $1\rightarrow 2$ be allowed in the dipole approximation. The levels 2 and 3 have like parity, but the presence of collisions with the buffer-gas atoms makes transitions between them possible. The balance equations for the level-population densities n_1, n_2, and n_3 of such a system are of the form

$$\begin{aligned} dn_1/dt&=-W_{13}(n_1-n_3)+n_2/\tau_{21}+n_3/\tau_{31}+W_{12}(n_2-n_1),\\ dn_2/dt&=-W_{12}(n_2-n_1)-n_2/\tau_{21}-n_2/\tau_{23}+n_3/\tau_{32},\\ dn_3/dt&=W_{13}(n_1-n_3)+n_2/\tau_{23}-n_3/\tau_{32}-n_3/\tau_{31},\end{aligned} \tag{1}$$

where $n = n_1 + n_2 + n_3$ is the density of the active atoms, W_{ij} is the probability of the stimulated single-photon transition $i\rightarrow j$, and τ_{ij} is the time of spontaneous or collisional transition $i\rightarrow j$.

Solving the system (1) for the stationary case we obtain expressions for the population differences Δ_{ij} of the levels i and j. Recognizing that $W_{13} = \sigma_{13}I$, where σ_{13} is the cross section of the single-photon transition $1\rightarrow 3$ and depends on the frequency of the exciting radiation,[9] while I is the intensity of the exciting radiation, we can express the dependence of Δ_{21}, Δ_{23}, Δ_{31} on I in the form

$$\Delta_{21}=\frac{aI-b}{cI+d},\quad \Delta_{23}=\frac{aI+e}{cI+d},\quad \Delta_{31}=-\frac{b+e}{cI+d}, \tag{2}$$

where

$$\begin{aligned} a&=\tau_{31}\tau_{32}(1/\tau_{32}-1/\tau_{23}-1/\tau_{21})\sigma_{13},\\ b&=\tau_{32}/\tau_{21}+\tau_{32}/\tau_{23}+\tau_{31}/\tau_{21},\\ c&=\tau_{31}\tau_{32}(3W_{12}+2/\tau_{23}+2/\tau_{21}+1/\tau_{32})\sigma_{13},\\ d&=2W_{12}(\tau_{31}+\tau_{32})+W_{12}\tau_{31}\tau_{32}/\tau_{23}+\tau_{31}/\tau_{21}+\tau_{32}/\tau_{21}+\tau_{32}/\tau_{23},\\ e&=\tau_{31}\tau_{32}(1/\tau_{32}+1/\tau_{31}-1/\tau_{23})W_{12}.\end{aligned} \tag{3}$$

It can be seen from (3) that b, c, and d are positive. The quantity e is also positive, for we always have $\tau_{32} < \tau_{23}$.[9] The sign of a is determined by the relation between τ_{32}, τ_{23}, τ_{21}. At $a < 0$ the quantity Δ_{21} is always negative, while $\Delta_{23} < 0$ under the condition $|a|I > e$. At $a > 0$ the quantity Δ_{23} is positive and Δ_{21} becomes larger than zero when the additional condition $aI - b > 0$ is satisfied. This points to the existence of an intensity threshold for the inversion of the populations of levels 2 and 1. It follows from Eqs. (2) that with increasing I the values of Δ_{21} and Δ_{23} tend to the same value a/c, i.e., the populations of levels 1 and 3 become equalized. We note that Δ_{31} is always less than zero.

The expressions for the dependences of Δ_{21} and Δ_{23} on the buffer-gas pressure P_b, obtained from (2) with allowance for the fact that τ_{23}^{-1} and τ_{32}^{-1} are proportional to P_b, are too long to be presented here. An analysis of these dependences shows that Δ_{21} and Δ_{23} becomes positive starting

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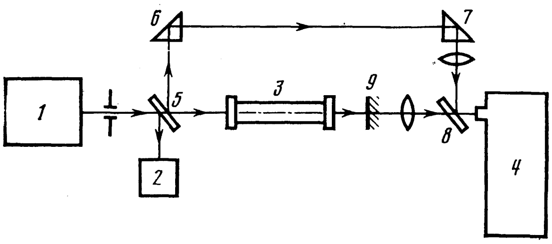

FIG. 1. Diagram of experimental setup. 1—dye laser, 2—laser-energy meter, 3—cell with potassium and buffer gas, 4—DFS-8 spectrograph, 5, 8—mirrors ($R = 50\%$), 6,7—prisms, 9—mirror ($R = 100\%$).

with a certain threshold value of the buffer-gas pressure. With increasing P_b, both Δ_{21} and Δ_{23} tend to certain constant values.

II. The experiments were performed with the setup shown schematically in Fig. 1. The emission of dye laser 1 was directed to cell 3 of length $l = 20$ cm filled with a mixture of potassium vapor and helium buffer gas. The scattered radiation was investigated with spectrograph 4 (DFS-8). Mirrors and prisms 6–9 were installed in the setup to investigate the directivities and the gains at the SERS and IRE.

The dye laser was excited with a ruby laser. We used a solution of DD1 dye in glycerin at a concentration $\sim 10^{-4}$ mol/1. The selective element of the dye-laser cavity was a diffraction grating with a two-prism beam expander. The laser emission frequency was tunable in the range 12900–13100 cm^{-1}. The emission energy was $\sim$ 1 mJ at a pulse duration 30–40 nsec and a spectral line width $\lesssim 0.5$ cm^{-1}. The dye-laser emission was linearly polarized, and the beam divergence was $\sim 10^{-2}$ rad.

Provision was made for independent variation of the potassium-vapor and buffer-gas pressures. The potassium-vapor pressure was varied between 10^{-4} and 10^{-2} Torr, and that of the buffer gas from 10 to 600 Torr.

III. We determined in the experiment the ranges of the exciting-radiation intensity and frequency and of the potassium-vapor and buffer-gas pressures when the scattering spectrum revealed the presence of radiation at the frequencies of the IRE (D_1 line) and of the SERS Stokes component. By adjusting the experimental conditions it was possible to obtain separately one of these emissions (see Figs. 3–5 below). Under the conditions of our experiment, no IRE was observed on the spectral line D_2 ($\Delta_{31} < 0$, see Sec I). The emission of the SERS anti-Stokes component, connected with $4P_{3/2} \to 4P_{1/2}$ transition,[10] could be observed at low values of the buffer gas (less than ~ 1 Torr).

The necessary conditions for the observation of IRE and SERS are $\Delta_{21} > 0$ and $\Delta_{23} > 0$. These conditions, according to Sec. I, are satisfied only if $a > 0$, i.e.,

$$\tau_{32}/\tau_{23} < 1 - \tau_{32}/\tau_{21}.$$

The last inequality can be satisfied under the condition $\tau_{32}/\tau_{21} < 1$, i.e., at a sufficiently high buffer-gas pressure whose value can be estimated by using the known formula $\tau_{32}^{-1} = N_b\, \sigma_{32} v$, where N_b is the density of the buffer-gas atoms, v is the mean squared relative velocity of the potassium and helium atoms, and σ_{32} is the cross section for the collisional transition $4P_{3/2} \to 4P_{1/2}$. Under the conditions of our experiment $v = 1.7 \times 10^5$ cm · sec^{-1} (the temperature of the medium is $T = 450$ K), $\tau_{21} \sim 3 \times 10^{-8}$ sec (Ref. 11), and $\sigma_{32} \sim 4 \times 10^{-15}$ cm^2 (Ref. 12), whence $N_b = 6 \times 10^{16}$ cm^{-3}. The necessary condition for the onset of IRE and SERS is thus satisfied at $P_b \gtrsim 3$ Torr, in full agreement with our experimental data.

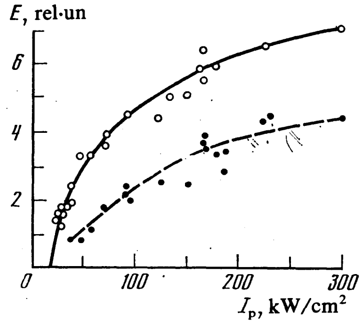

FIG. 2. Dependences of IRE and SERS energies on the exciting-radiation intensity I_p. Curves—calculation, light circles—experimental values of the IRE energy, dark circles—experimental values of SERS energy. Exciting-radiation frequency $\omega_p = 13039$ cm^{-1}, buffer-gas pressure $P_b = 500$ Torr, potassium-vapor pressure $P(K) = 1 \times 10^{-3}$ Torr.

Figure 2 shows the dependences of the IRE and SERS emission energies on the exciting-radiation intensity. The dependences obtained are well approximated by a function of the form $E = E_0 \exp(\sigma \Delta l)$, where E_0 is the initial energy, σ is the cross section of the corresponding transition, Δ is the population difference calculated from Eq. (2), and l is the length of the active zone. This function is a solution of the propagation equation under the assumption that Δ is constant along the propagation direction of the radiation in the medium.

For the case cited, the efficiency of conversion into IRE and SERS is $\sim 1\%$ in energy for high exciting-radiation intensities, and $\sim 10\%$ for low ones.

Under the conditions of our experiment the IRE energy is radiated symmetrically along and counter to the propagation direction of the exciting radiation. The SERS is emitted along in the exciting-radiation propagation direction. The divergence of both emissions is $\sim 10^{-2}$ rad.

The observed IRE emission was depolarized, while the SERS polarization coincided with that of the exciting radi-

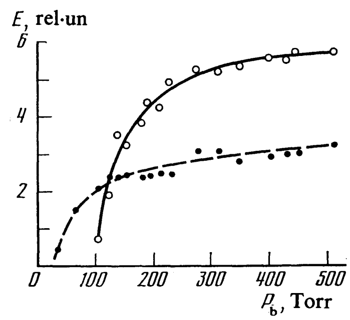

FIG. 3. Experimental dependences of the IRE and SERS energies on the buffer-gas pressure. Solid curve—IRE, dashed—SERS. Frequency of exciting radiation $\omega_p = 13041$ cm^{-1}, $P(K) = 1 \times 10^{-3}$ Torr, $I_p = 300$ kW/cm^2.

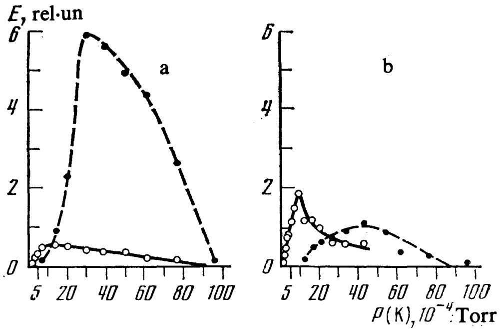

FIG. 4. Experimental dependences of the IRE and SERS energies on the potassium vapor pressure at different buffer-gas pressures: a—$P_b = 190$ Torr, exciting-radiation frequency $\omega_p = 13048\ \mathrm{cm}^{-1}$; b—$P_b = 500$ Torr, $\omega_p = 13050\ \mathrm{cm}^{-1}$. $I_p = 300\ \mathrm{kW/cm}^2$. Solid curve—IRE, dashed—SERS.

ation. The depolarization of the IRE is due to the fact that this process starts out from depolarized spontaneous resonant emission, and the gain is the same at all polarizations. The reason why the SERS is polarized is that the gain of this emission is a maximum when the polarization coincides with that of the exciting radiation.

The experimental dependences of the IRE and SERS energies on the buffer gas pressure (Fig. 3) show that the IRE and SERS processes have definite buffer-gas-pressure thresholds and reach constant values at high buffer-gas pressures. This agrees with the deductions cited above concerning the dependences of Δ (and hence of E) on the buffer-gas pressure. The experimental values of the threshold pressures of the buffer gas also agree well with the calculated ones.

Figure 4 shows the experimental dependences of the IRE and SERS energies on the potassium-vapor pressure. In the cases cited, when the potassium vapor pressure is raised to $\sim 10^{-3}$ Torr the IRE energy increases to the maximum. The SERS emission appears at practically the same potassium-vapor pressure and its energy reaches a maximum at $P(\mathrm{K}) = 4\times 10^{-3}$ Torr. It can be seen from the presented relations that the ratio of the IRE and SERS energies changes with change of the buffer gas pressure. The decrease of the IRE and SERS energies with increasing potassium-vapor pressure is apparently due to the increase of the optical thickness of the absorbing layer.

An analysis of the frequency dependences of expressions (2) shows that the IRE and SERS energies should increase as the exciting-radiation frequency approaches resonance with the $4S_{1/2}\rightarrow 4P_{3/2}$ transition, owing to the increased absorption cross section σ_{13} (see Sec. I). As seen from the experimental plots of Fig. 5, this is indeed the case. The maximum values of the IRE and SERS are reached not at exact resonance: for IRE and SERS the maximum points are shifted respectively towards higher and lower frequencies. This asymmetry can be due to the asymmetric broadening and shift of the resonant spectral lines in the presence of a buffer gas.[13,14] It can also be seen from Fig. 5 that the SERS energy, in contrast to the IRE, changes little with changing pressure of the buffer gas in the entire range of variation of the exciting-radiation frequency.

The experimental investigations have shown that when

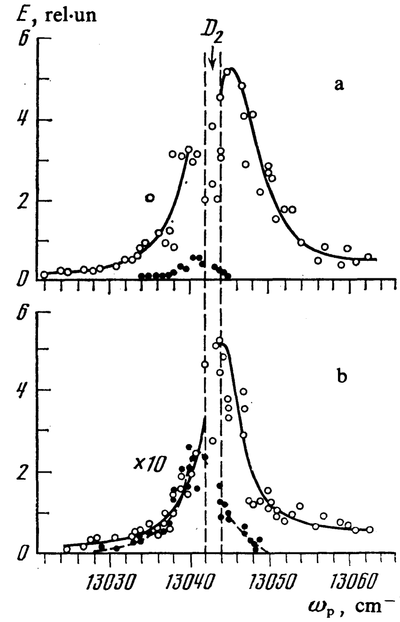

FIG. 5. Experimental dependences of the IRE and SERS energies on exciting-radiation frequency at different values of the buffer gas pressure. a—$P_b = 500$ Torr, b—$P_b = 190$ Torr, $P(\mathrm{K}) = 1\times 10^{-3}$ Torr, $I_p = 300\ \mathrm{kW/cm}^2$. Solid curve—IRE, dashed—SERS. In the separated frequency region the spectral line of the exciting radiation overlaps the D_2 absorption line.

the potassium vapor is excited by radiation with frequency close to the resonant $4S_{1/2}\rightarrow 4P_{1/2}$ transition neither SERS nor IRE is observed. The absence of IRE in this case is natural, since the exciting radiation can only equalize the populations of levels 1 and 2. The absence of SERS is attributed to the abrupt decrease of the cross section of the process, for in the case the virtual intermediate level is separated from the nearest resonant level by an amount $\sim E_3 - E_2$.

The results cited show that a mixture of alkali metal vapor and inert gas, with suitably chosen parameters, can serve as a convenient active medium for laser emission at an atomic resonant frequency.

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