Investigation of the dynamics of the development of nonlinear resonance processes in potassium vapor

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An experimental investigation of the temporal characteristics of the radiation of a resonant medium has been carried out with a time resolution 10^{-10} sec. It was demonstrated that, when a high-intensity exciting resonance radiation interacts with potassium vapor, the frequency of the exciting radiation in the forepart of the pulse changes. The frequency shift is explained by the phase modulation that occurs during the inverse population of the medium. A frequency shift, which is also explained by the phase modulation, was observed in the forepart of the stimulated-electron-Raman-scattering (SERS) pulse. It was demonstrated that the intensification of the three-photon scattering process occurs during the counter propagation of the exciting-radiation wave primarily because of the duration of the process.

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1. INTRODUCTION

Much attention has been given in recent years to the investigation of the interaction of high-intensity resonance radiation with the vapors of the alkali metals,[1] in particular, potassium. This is connected with the high nonlinearities that manifest themselves in resonance interactions, and also, because of the low density of vapors, with the simplification that can be made in the theoretical analysis of the experimentally observable nonlinear effects. The nonlinear processes that occur in potassium vapor near a resonance—self focusing, spectrum broadening, pulse-shape alteration, stimulated electron Raman scattering (SERS), three-photon scattering, etc.—have been quite well studied both theoretically and experimentally.[2–11] Nevertheless, the development in time of these processes has at present virtually not been investigated.

The present paper is devoted to the experimental study of the development in time of the processes of exciting-radiation-spectrum broadening, three-photon scattering, and SERS in potassium vapor.

2. EXPERIMENTAL SETUP

The investigation of the dynamics of the development of the phenomena arising during the passage of intense radiation through a resonance medium was carried out with the apparatus shown in Fig. 1. The radiation of a Q-switched, 10–20-MW, ruby laser 1 was directed at a cuvette with an organic liquid 2. The stimulated-Raman-scattering pulse from the organic liquid, after passing through a cuvette with potassium vapor 5, hit the slit of a spectrograph 6. The mode composition of the radiation was not controlled. For comparison, part of the radiation was diverted to another spectrograph 7 before it entered the cuvette with the potassium vapor. The focal planes of the cells of both spectrographs were projected onto the photocathode of a photoelectronic detector 8, which provided a sweep of the spectrum in time. On the obtained spectrochronograms, the directions of sweep over time and over the spectrum were mutually perpendicular. The choice of the organic liquid was determined by the requirement that the frequency of the SRS radiation be close to the frequency of the resonance transition $4S_{1/2}-4P_{3/2}$ (13 043 cm^{-1}) in potassium. For the excitation we used the second Stokes SRS component in chloroform with frequency 13 066 cm^{-1}, i.e., the detuning was 23 cm^{-1}. To trigger the photoelectronic detector, and to measure the energy of the exciting radiation, we diverted portions of the corresponding radiations to the photocell and the calorimeter 4. The ruby laser radiation was blocked by the filter 3. The cuvette with potassium was 30 cm long, and was heated up to a temperature of 250–340 °C, which corresponds to a saturated potassium-vapor density of 10^{15}–2×10^{16} at/cm^3. As the dispersive element, we used ISP-51 spectrographs with different cells, so that the dispersion on the screen of the image-converter tube was 26 cm^{-1}/mm for the main channel and 740 cm^{-1}/mm for the comparison channel. The duration of a sweep of the spectra was equal to 1.8×10^{-9} sec per mm of screen. The total pulse duration was measured at the 10^{-7}-J/cm^2 level,[12] and was found to be 30–50 nsec for the laser pulse and 10–25 nsec for the pulse of the second Stokes SRS component in chloroform. The experiments were performed with the exciting radiation focused in the potassium vapor.

Besides the above-described "direct" scheme, we also used for the study of the many-photon processes that occur in the case of counter propagation of the exciting-radiation waves the "backward" scheme,[13] in which case we introduced the mirrors 9, 10, and 11 into the apparatus. In this case the radiation that passed through

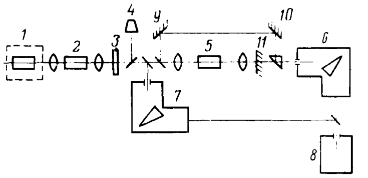

FIG. 1. Diagram of the experimental setup: 1) ruby laser with Q switching; 2) cuvette with organic liquid and of length $l=20$ cm; 3) FS-7 filter; 4) calorimeter; 5) cuvette with potassium; 6), 7) ISP-51 spectrographs; 8) FER-2 photoelectronic detector; 9)–11) 100-percent dielectric mirrors.

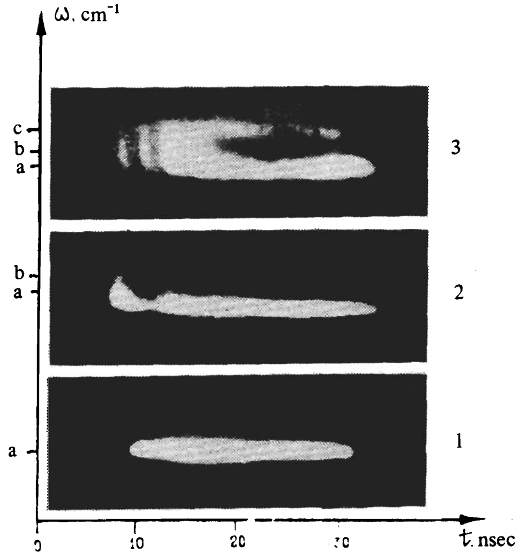

FIG. 2. Spectrochronograms of the stimulated processes in potassium vapor in the case of excitation by resonance radiation. The "direct" scheme. Temperature of the potassium vapor: 1) 190 °C, 2) 300 °C, 3) 340 °C; a) incident radiation, b) the three-photon scattering radiation, c) the SERS radiation.

the potassium vapor and was reflected from the mirror 11 met in the cuvette with the remaining part of the pulse and was further directed by the mirrors 9 and 10 to the slit of the spectrograph 6.

3. EXPERIMENTAL RESULTS

A. The direct scheme

During the passage of the focused exciting radiation through the potassium vapor under the conditions of our experiment, we observed, starting from a vapor density of 10^{15} at/cm^3, the variation in time of the spectral composition of the transmitted radiation. Especially strong was the variation of the frequency of the forepart of the incident pulse: It increased discontinuously within a time less than 10^{-10} sec by a value roughly equal to the detuning and then decreased relatively slowly over a period of 1–2 nsec to the former frequency. The duration of the transition to the fundamental frequency increased with the growth of the vapor density. Similar shifts were observed at other parts of the pulse at densities of 10^{16}–2×10^{16} at/cm^3. In a number of cases a separate pulse of length 10^{-10} sec can be distinguished on the obtained spectrochronograms at the three-photon scattering frequency. The SERS line, whose width increased with increasing vapor density from 2 to 8 nsec, appeared somewhat later than was expected. The temporal structure of the SERS pulse essentially repeated the structure of the exciting-radiation pulse. The lag (frontwise) of the SERS behind the exciting radiation also increased with the growth of the potassium-vapor density. The frequency of the SERS line varied in time. At the beginning of the pulse it was close to the incident frequency, but it subsequently increased smoothly to the fundamental value of 13 124 cm^{-1} (see Fig. 2).

B. The backward scheme

The laws indicated in Sec. 1 as governing the development of the processes essentially remain valid in the case of the counter propagation of the exciting-radiation waves, especially at relatively low potassium-vapor densities. Again, starting from a density of 10^{15} at/cm^3, the frequency of the forepart of the incident pulse shifts discontinuously to the three-photon scattering line and then smoothly proceeds to the fundamental line. The duration of the transition varies from 1.5 to 4 nsec and also increases with increasing vapor density. At the same time as the vapor density increases from 10^{15} to 10^{16} at/cm^3 the duration of the pulse at the frequency of the exciting radiation drops from 20 to 10 nsec at the expense of the pulse's forepart, whose energy is converted into the frequency of the three-photon scattering and SERS (see Fig. 3).

In contrast to the direct scheme of the experiment, the duration of the pulse of the three-photon process increases, as the vapor density increases, from 2 to 20 nsec, and becomes at high densities (10^{16} at/cm^3) comparable to the duration of the exciting-radiation pulse. This shows that the intensification, observed, for example, in[8], of the three-photon process occurs largely because of the duration of the process.

As in the preceding case, in the case of the counter propagation of the exciting-radiation waves, the SERS line appeared at a density of 10^{15} at/cm^3 virtually simultaneously with the three-photon scattering line.

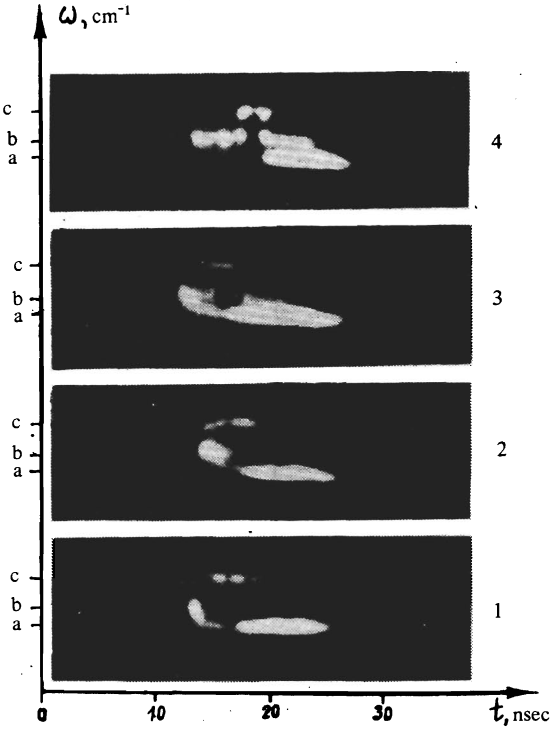

FIG. 3. Spectrochronograms of the stimulated processes in potassium vapor in the case of excitation by resonance radiation. The "backward" scheme. Temperature of the potassium vapor: 1) 305 °C, 2) 315 °C, 3) 320 °C, 4) 340 °C; a) incident radiation, b) the three-photon scattering radiation, c) the SERS radiation.

Upon further increase of the vapor density, the SERS line appeared later than the three-photon scattering line. For a vapor density of 2×10^{16} at/cm^3 the magnitude of this lat attained a value of 8 nsec. The duration of the SERS pulse was between 2 and 10 nsec. The temporal structure of the SERS line essentially repeated the structure of the exciting-radiation pulse. The relative intensity of the SERS radiation was higher in the backward scheme than in the direct scheme.

4. DISCUSSION OF THE OBTAINED RESULTS

The variations in time of the frequency of the exciting-radiation spectrum that were observed in both schemes of the experiment, i.e., in both the direct and backward schemes, in the case when the radiation was focused in the potassium vapor, as well as the theoretical estimates lead to the following conclusion about the course of the process.

Upon the incidence of the resonance radiation on the potassium vapor, the three-photon process, which overpopulates the $4P_{3/2}$ level relative to the ground level, very quickly develops—according to our estimates (based on[14]), over a period of 10^{-11} sec. The process, inverse to the three-photon process, which could empty the upper level cannot occur, since under the conditions of our experiment, because of the small difference between the group velocities of the incident and three-photon radiations, virtually no photon exists at the three-photon scattering frequency in the volume with the inverse population. The duration of the three-photon process is less than the $4P_{3/2}$ level's relaxation time determined with allowance for collisions.[15] In view of this, the three-photon scattering process quickly ceases.

The rapid pumping of the energy of the exciting radiation into the three-photon process will lead to the formation of a steep leading edge in the exciting-radiation pulse. That the steep leading edge arises as a result of the interaction with the potassium vapor is attested by the fact that, although the shape of the exciting pulse changed from pulse to pulse, the magnitude of the frequency shift remained constant, other conditions of the experiment being equal. Under these conditions the process of phase modulation plays an appreciable role in the variation of the frequency.

Let us consider the phase of the forward wave in the form

$$\psi=\omega t-\frac{\omega}{c}nx;$$

then the phase-modulation-induced frequency change

$$\delta\omega=-\frac{\omega}{c}x\frac{\partial n}{\partial t}.$$

Thus, the sign and magnitude of the frequency shift depend on the sign and magnitude of the time derivative of the refractive index n. Using the expression given in[16] for the refractive index

$$n=1+\frac{2\pi\Delta|d|^2}{\hbar\varepsilon\sqrt{1+\xi}},$$

we find

$$\delta\omega=\frac{\pi\omega|d|^2x}{c\hbar\varepsilon}\Delta(1+\xi)^{-3/2}\frac{\partial\xi}{\partial t},$$

where $\xi=4|dE|^2/\hbar^2\varepsilon^2$ is a dimensionless parameter of the intensity; d is the matrix element of the $4S_{1/2}-4P_{3/2}$ transition; E is the field intensity, $\varepsilon=\omega_0-\omega$, ω_0 and ω being the atomic-transition and incident-radiation frequencies; $\Delta=N_1-N_2$ is the difference between the populations of the $4S_{1/2}$ and $4P_{3/2}$ levels; x is the length of the cuvette; and c is the velocity of light.

Since the derivative $\partial\xi/\partial t\sim\partial I/\partial t$ (I is the intensity density) is positive (i.e., since $\partial\xi/\partial t>0$) at the leading edge of the pulse, the shift, obtained in our experiment, of the exciting frequency toward the region of higher frequencies can occur only when $\Delta<0$. This confirms our assumption that an overpopulated state is formed between the $4S_{1/2}$ and $4P_{3/2}$ states as a result of the three-photon process.

A numerical estimate of the frequency shift for $\omega=13066$ cm^{-1}, $|d|^2=3.4\times10^{-35}$ cgs esu, $|\Delta|\sim10^{16}$, $\varepsilon=-23$ cm^{-1}, $\xi\sim1$, and $\partial\xi/\partial t\sim10^{11}$ gives $\delta\omega\sim10^{12}$ Hz ≈10 cm^{-1}, which agrees in order of magnitude with the experiment.

The observed smoother transition of the scattering frequency to the fundamental frequency as the vapor density was increased was due, apparently, to the decrease of the rate of damping of the three-photon process.

No frequency shift was observed by us at the remaining part of the exciting pulse, owing to its small slope. The observed frequency shift of the leading edge of the SERS pulse can also be explained by the phase-modulation process. SERS in a three-level system has been theoretically investigated in[17], where an expression is given for the SERS enhancement ratio. Using this formula and the Kramers-Krönig relation, we obtain for the refractive index n at the SERS frequency the expression:

$$n=1-\frac{\pi|d|^2\Delta\varepsilon^2\xi(\sqrt{1+\xi}+1)}{8\hbar\varepsilon_1{}^2\Gamma\cdot4\sqrt{1+\xi}}.$$

The formula has been derived for SERS detunings of the order of the width, Γ, of the $4P_{3/2}-4P_{1/2}$ transition. In our case the sign of the detuning with allowance for the Stark shift should be taken to be negative. The frequency shift due to the phase modulation will have, in this case, the form

$$\delta\omega=\frac{\pi\omega|d|^2x\varepsilon^2\Delta}{32c\hbar\varepsilon_1{}^2\Gamma}\left[\frac{2(1+\xi)^{3/2}+\xi+2}{2(1+\xi)^{3/2}}\right]\frac{\partial\xi}{\partial t},$$

where $\varepsilon_1=\omega_0-\omega_{\mathrm{rem}}=81$ cm^{-1} and the notation for the remaining quantities has the same meaning as before. These expressions have been derived under the conditions that $\Delta_1=N_1-N_3=0$ (N_3 is the population of the $4P_{1/2}$ level) and $\Delta<0$.

The substitution of the numerical values of the quantities into the formula gives for the leading edge of the pulse $\delta\omega<0$ and $|\delta\omega|\sim10^{12}$ Hz ~10 cm^{-1}, which agrees with the experimental result given above.

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Rotational spectrum of diatomic molecules in the field of an intense electromagnetic wave

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Rotational spectra of diatomic heteronuclear molecules in the field of an external electromagnetic wave are investigated. Explicit expressions are obtained for the dynamic Stark effect in the vibrational-rotational levels of the molecule in a nonresonance field. A discussion is given of the resonance Stark effect, in which the frequency of the wave is close to the frequency of a vibrational transition. It is shown that in a resonance field with a field strength $F_0\sim(10^5–10^6)$ V/cm a radical restructuring of the rotational spectra of the molecule occurs. It is found that in still more intense fields there is an orientation of the molecules (as the result of the resonance Stark effect) in directions perpendicular to the field F_0.

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1. INTRODUCTION

It is well known that the action of a weak constant or alternating (nonresonance) electric field on a molecule produces only small shifts of the vibrational-rotational levels. The magnitude of these shifts can be determined by means of the usual formulas of the static[1–4] or dynamic[3–5] Stark effects in terms of perturbation theory.

Explicit expressions for the quadratic Stark shift of molecular levels in an alternating field due to the vibrational and rotational motions of the nuclei in the molecule were obtained recently by Braun and Petelin.[5] In Sec. 2 of the present article we discuss the dynamic Stark effect in second-order perturbation theory with inclusion of the electronic motion in addition to the vibrational and rotational motions.

The most interesting case is that in which the external field cannot be considered small, i.e., when the Stark shift becomes comparable with the distance between the rotational sublevels. The restructuring of the rotational spectrum in a strong constant field has been discussed by several authors.[6–9] In the case of an alternating field, in the nonresonance case the interaction ceases to be small at $d_0F_0\sim\hbar\omega_e$, where d_0 is the characteristic value of the dipole moment of an electronic transition, F_0 is the electric field strength, and ω_e is the vibrational frequency. In the resonance case $\omega\approx\omega_e$, a substantial rearrangement of the rotational spectrum arises at much lower fields, when $V_0\equiv d_1F_0\sim B_e$, where d_1 is the characteristic value of the dipole moment of a vibrational transition ($d_1\sim(m/M)^{1/4}d_0$, m is the electron mass, and M is the reduced mass of the nuclei) and B_e is the rotational constant of the molecule.

In Sec. 3 we discuss the problem of the rotational spectrum of a diatomic heteronuclear molecule in a linearly polarized electromagnetic wave of not too high