SOME NONLINEAR OPTICAL EFFECTS IN POTASSIUM VAPOR

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Some nonlinear effects (dependence of the group velocity on the intensity, phase modulation, pulse shape distortion, appearance of combined lines due to multiphoton interaction processes) are investigated theoretically near the $4P_{3/2}$–$4S_{1/2}$ resonance of the potassium atom. Broadening of the spectral line of the second Stokes component of stimulated Raman scattering of chloroform is observed on passage through a cell containing potassium at a saturated vapor pressure of 0.05–1.7 mm Hg. Three-photon and five-photon scattering was observed under the same experimental conditions.

1. INTRODUCTION

APPRECIABLE attention has been paid recently to nonlinear optical phenomena in gases. The narrowness of the energy levels, the possibility of smooth variation of the density, pressure, resonance detuning, and the relative simplicity and clarity of the theoretical interpretations make gases interesting objects of study in nonlinear optics.

In[1,2] they succeeded, for the first time, in observing the Stark phenomenon in a resonant situation for frequencies in the optical band. In[3-5] observation was reported of stimulated electron Raman scattering with excitation of the $4P_{3/2}$ level of potassium atoms. In[5,6], stimulated emission was obtained from different levels of the potassium atom. In[7] are presented results of observation of three-photon interaction in rubidium-atom vapor.

The indicated nonlinear effects have a clearly resonant character. Some nonlinear effects (self-focusing, broadening of the spectral lines due to phase modulation, etc.) resulting from the nonlinear part of the refractive index and investigated in relatively great detail in liquid and solids far from resonance, have not been investigated under resonant conditions. The nonlinearity of such media can be come appreciable as a result of the closeness of the resonance, in spite of the low density of the active atoms.

We report in this paper the results of a theoretical and experimental investigation of certain nonlinear effects (the dependence of the change of phase and of the group velocity on the intensity, the broadening of the spectrum, the appearance of new combination frequencies as a result of multiphoton interaction processes) near the resonance $4P_{3/2}$–$4S_{1/2}$ of the potassium atom.

2. THEORETICAL PART

We make the following assumption concerning the resonance detuning $\epsilon = \omega_0 - \omega'$ (ω_0—resonant frequency of the atoms, ω'—frequency of incident wave), the pulse duration T, and the irreversible relaxation time τ: $\epsilon \gg T^{-1} \gg \tau^{-1}$. It is then possible to neglect relaxation processes and the use for the amplitude of the vector potential a(t, x) and the transition current $\rho(t, x)$ the following equations (see[8]):

$$\frac{\partial a}{\partial x} + \frac{1}{c}\frac{\partial a}{\partial t} = -p\rho, \tag{1}$$

$$\frac{\partial \rho}{\partial t} + i\epsilon\rho = a\sqrt{1 - q^2|\rho|^2}. \tag{2}$$

Here $p = 2\pi|M|^2N/\omega' c\hbar$, $q = 2|M|/c\hbar$, M is the matrix element of the transition, and N is the density of the atoms. The current density ρ determined from (2) is characterized both by rapid variations (with frequency ϵ) and by slow ones (with the frequency of variation of the quantity a). The modulation of the current with frequency ϵ leads to emission of new lines with frequencies $\omega' \pm \epsilon$, $\omega' \pm 2\epsilon$, etc. Since the spontaneous effects are not taken into account, the probability of emission of these new lines will be different from zero if the incident radiation has nonzero components at these frequencies. If the spectral width of the incident radiation is $\Delta\omega \ll \epsilon$, then the probability of emission of these lines is small and rapid variations of the current can be neglected (this is equivalent to the condition $|\partial\rho/\partial t| \ll |\epsilon\rho|$).

A rigorous mathematical proof of the possibility of neglecting $\partial\rho/\partial t$ is based on the changeover to spectral components of a and ρ in Eqs. (1) and (2). We shall demonstrate this, by way of an example, for the linear case

$$a(t, x) = \int_{-\infty}^{+\infty} F_0(\omega) \exp\left\{-i(\omega - \omega')\left(t - \frac{x}{c}\right) + i\frac{px}{\varepsilon - (\omega - \omega')}\right\} d\omega. \tag{3}$$

Here $F_0(\omega)$ is the emission spectrum incident on the boundary x = 0. Since $F_0(\omega)$ has a sharp maximum at $\omega = \omega'$ with width $\Delta\omega \ll \epsilon$, we can put in (3) $|\omega - \omega'| \ll \epsilon$ and the expression in the argument of the exponential can be expanded in powers of $|\omega - \omega'|/\epsilon \ll 1$. Confining ourselves to the first two terms of the expansion, we get

$$a(t, x) = f\left(t - \frac{x}{c} - \frac{px}{\varepsilon^2}\right) \exp\left(i\frac{px}{\varepsilon}\right), \tag{4}$$

where f(t) = a(t, 0). This result agrees with (1) and (2) if we regard $\partial\rho/\partial t$ in (2) as a small perturbation. The next terms of the expansion in (3) correspond to the dispersion effects of the spreading of the momen-

tum and appear at sufficiently x ($x \gtrsim x_0 = \epsilon^3/p\Delta\omega$—in real cases x_0 is of the order of several meters.

Thus, regarding $\partial\rho/\partial t$ in (2) as a small perturbation, we get

$$\frac{\partial a}{\partial x}+\frac{1}{v}\frac{\partial a}{\partial t}=\frac{ip}{\varepsilon}\frac{a}{(1+q^2|a|^2/\varepsilon^2)^{1/2}}, \tag{5}$$

where

$$\frac{1}{v}=\frac{1}{c}+\frac{p}{\varepsilon^2(1+q^2|a|^2/\varepsilon^2)^{3/2}}. \tag{6}$$

The solution of this equation is of the form

$$a=f(\tau)\exp\left[\frac{ipx}{\varepsilon(1+q^2|f(\tau)|^2/\varepsilon^2)^{1/2}}\right], \tag{7}$$

where

$$\tau=t-x\left[\frac{1}{c}+\frac{p}{\varepsilon^2(1+q^2|f(\tau)|^2/\varepsilon^2)^{3/2}}\right]. \tag{8}$$

Formula (6) gives the group velocity of propagation of a pulse in a medium with allowance for the intensity effect, and (7) gives the distortions of the phase and of the amplitude of the pulse. The broadening of the spectrum due to these effects has been considered in[9]. We present here only the expression for the spectral width at a distance x in the case $px/\epsilon \gg 1$ (under the conditions of the present experiment this takes place):

$$\Delta\omega(x)=\left[(\Delta\omega)^2+\left(\frac{px}{\varepsilon T}\right)^2\frac{\xi^2}{(1+\xi)^3}\right]^{1/2}, \tag{9}$$

where $\xi = q^2|f(0)|^2/\epsilon^2$. For allowed atomic transitions and for pulses with $T \sim 10^9$ sec, $\Delta\omega \sim 10^{11}$ sec^{-1}, and $\epsilon \sim 10^2$ sec^{-1}, the additional broadening will be noticeable when x is of the order of several centimeters.

We now proceed to consider the effects of current modulation. For simplicity we confine ourselves to the case of weak nonlinearity $q^2|a|^2/\epsilon^2 \ll 1$ and change over in (1) and (2) to spectral components of the field $F(\omega, x)$:

$$i(\omega_0-\omega)\frac{d}{dx}F(\omega,x)+pF(\omega,x) = \frac{pq^2}{2}\int\int\frac{d\omega_1 d\omega_2}{(\omega_0-\omega_1)(\omega_0-\omega_2)}F(\omega_1,x)F^*(\omega_2,x)F(\omega+\omega_2-\omega_1,x). \tag{10}$$

We assume that the incident radiation consists of a strong narrow part (for simplicity, monochromatic) and a weak continuous background, i.e.,

$$F(\omega, 0)=\gamma_0\delta(\omega-\omega')+\beta_0(\omega). \tag{11}$$

It is easy to verify that $F(\omega, x)$ has the same form

$$F(\omega, x)=\gamma(x)\delta(\omega-\omega')+\beta(\omega, x). \tag{12}$$

If we confine ourselves to terms linear in β, then we obtain for β a second-order equation with constant coefficients. The determinant of the equation for the characteristic roots is given by the expression

$$\delta=\frac{p^2}{\varepsilon^2}(1-\eta)^2\frac{z^2}{(1-z^2)^4}\left(1-\frac{2\eta}{1-\eta}-z^2\right)\left(z^4-z^2+\frac{2\eta}{1-\eta}\right), \tag{13}$$

where $z = (\omega-\omega')/\epsilon$, $\eta = q^2|\gamma_0|^2/2\epsilon^2$. For frequencies for which $\delta(z) > 0$, amplification is possible. It follows from (13) that this occurs in the following regions (at $\eta \ll 1$):

$$\text{I.}\quad 0<z^2<\frac{2\eta}{1-\eta}+\frac{1}{2}\frac{\eta^2}{(1-\eta)^2}, \tag{14}$$

$$\text{II.}\quad 1-\frac{2\eta}{1-\eta}-\frac{1}{2}\frac{\eta^2}{(1-\eta)^2}<z^2<1-\frac{2\eta}{1-\eta}. \tag{15}$$

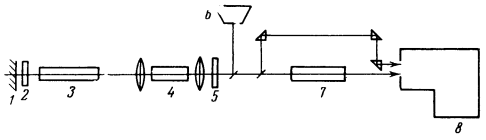

FIG. 1. Diagram of experimental setup: 1 – mirror, R = 99%; 2 – passive shutter, 3 – ruby rod, 4 – cell with organic liquid, 5 – FS-7, filter, 6 – calorimeter, 7 – cell with potassium vapor, 8 – spectrograph.

Region I is directly adjacent to the incident line, and the maximum amplification takes place here for $z^2 = \eta$. Region II corresponds to the frequencies $\omega \sim \omega_0$ and $\omega \sim 2\omega' - \omega_0$, and the maximum amplification occurs at $z^2 = 1 - 2\eta$. The amplification in region I can be ascribed to the decay of two photons $2\omega_0 = \omega_1 + \omega_2$, while that in region II to three-photon scattering, when the atom absorbs two quanta of frequency ω', emits a quantum of frequency $2\omega' - \omega_0$, and remains in an excited state. Owing to phase modulation, the frequency $2\omega' - \omega_0$ should be accompanied by a frequency ω_0, but the gain at the frequency ω_0 is less important, owing to the real absorption.

3. EXPERIMENTAL SETUP

An experimental investigation of the phenomena occurring when an intense light wave passes through a resonant medium was carried out with the apparatus shown in Fig. 1.

The laser operated in the giant-pulse regime with Q-switching with the aid of a passive shutter. The duration of the giant pulse was 25—30 nsec, and the power was on the order of 30 MW. The laser emission excited stimulated Raman scattering (SRS) in an organic liquid; this scattering was directed to the cell with vapor of atomic potassium, and then was incident on the spectrograph slit. The main laser radiation was eliminated by an FS-7 filter. Part of the incident beam was diverted to the upper part of the spectrograph slit, bypassing the cell with the potassium vapor. The experiments were performed both with the radiation focused in the cell, and without this focusing, with cells of different lengths from 2 to 100 cm. The temperature of the cell with the potassium was varied in the range from 250 to 350°C (the corresponding potassium saturated-vapor pressure ranged from 0.05 to 1.7 mm Hg).

The scattered radiation was registered in the direction of incident radiation with an ISP spectrograph (linear dispersion approximately 30 Å/mm in the region 13 000 cm^{-1}. In a number of experiments, a diffraction spectrograph DFS-13 was used (linear dispersion 4 Å/mm). The spectrograms were reduced by photographic photometry.

Certain time characteristics of the radiation were investigated with the aid of a coaxial photocell FEK-09 with an S1-11 oscilloscope. The time resolution of the system was ~5 nsec.

The choice of the medium for obtaining SRS was determined by the requirement that the frequency of the SRS be close to the frequencies of the resonant transitions $4P_{3/2}$—$4S_{1/2}$ (13 043 cm^{-1}) and $4P_{1/2}$—$4S_{1/2}$ (12 986 cm^{-1}) in potassium. We used the first SRS Stokes component of nitrobenzene with frequency

13 055 cm^{-1}, the second SRS Stokes component of chloroform with frequency 13 066 cm^{-1} (detuning 12 and 23 cm^{-1} for the $4P_{1/2}$–$4S_{1/2}$ transition), and the second Stokes SRS component of orthoxylol with frequency 12 940 cm^{-1} (detuning 46 cm^{-1} for the transition $4P_{1/2}$–$4S_{1/2}$).

4. BROADENING OF EMISSION SPECTRUM

When the second Stokes SRS component of chloroform was passed through a cell with potassium vapor, an appreciable broadening is observed in the spectra. The spectrogram (Fig. 2) shows clearly that the broadening has a resonant character. The first Stokes SRS component of chloroform present in the incident radiation does not experience a noticeable broadening. Besides the SRS of chloroform, we used as excitation sources also the SRS of nitrobenzene and orthoxylol. Comparison of the result of these investigations under identical experimental conditions shows that in the case of nitrobenzene there is appreciable broadening, but there is practically none for orthoxylol.

The observed broadening increase with increasing intensity of the incident radiation, and also with increasing potassium vapor pressure. Figure 3a shows the observed line contour of the emission of the second Stokes SRS component of chloroform after passing through potassium vapor at pressures 0.5 and 1.2 mm Hg (the decreasing intensities were approximately the same). For comparison we show the contour of the same line prior to the entry to the cell. On the long-wave side, the line contour is distorted and there is no wing, owing to the real resonant absorption in the potassium vapor. The observed half-width of the lines at half-height (measured from the undistorted half of the contour) for the emission of the second Stokes SRS component of chloroform prior to the entry to the cell is 1.5 cm^{-1}, and for the same line after the passage through the cell, under the indicated conditions, it is 2.0 and 4.4 cm^{-1} respectively (the spectral width of the gap was 0.3 cm^{-1}).

Figure 3b illustrates the dependence of the broadening of the incident intensity at constant vapor pressure of 0.5 mm Hg. The incident intensities differ by approximately a factor of 2. Such a spectrum broadening is attributed by us to the time modulation of the phase of the field on passing through the medium (see Sec. 2). Experimentally, the effective broadening of the spectral line as a result of the phase modulation is observed in nonresonant media by a number of authors[10-14]. A distinguishing feature of this experiment is the closeness of the resonance, as a result of which the effect of the broadening is observed in a gaseous medium at much lower densities (compared with[10-14]).

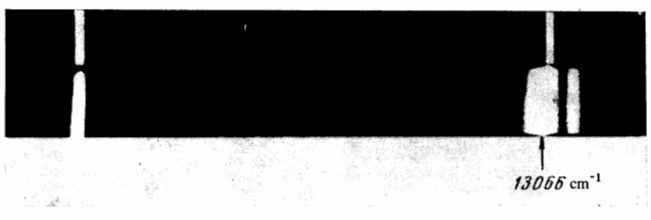

FIG. 2. Emission spectrogram of the first and second Stokes components of SRS of chloroform passing through potassium vapor (pressure 0.7 mm Hg). The spectrum of the incident radiation is shown for comparison.

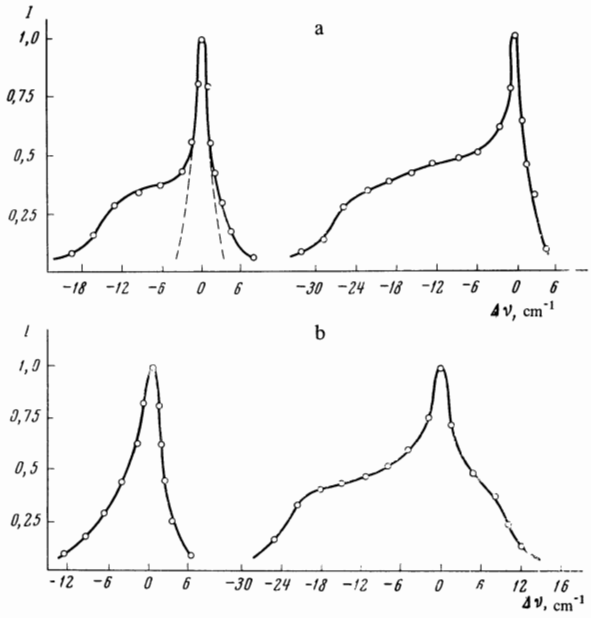

FIG. 3. Intensity distribution (relative units) in the second Stokes SRS component of chloroform: a – potassium vapor pressures 0.5 and 1.2 mm Hg, incident intensities approximately equal. The dashed line shows the contour of the incident line; b – vapor pressure 0.5 mm Hg, incident intensities differ by a factor of 2. The maximum intensities are normalized to unity.

In all the experiments, we monitored the total energy of the incident radiation, However, owing to the change in the space-time structure of the radiation from pulse to pulse, and also owing to the possible self-focusing in potassium vapor, the energy measurements do not reflect the picture of the real intensity, and therefore a detailed quantitative comparison with theory is impossible. Nonetheless, the main characteristics, such as the increase of the spectral width with increasing cell length, with increasing potassium vapor pressure and with decreasing intensity, as well as the order of magnitude of the involved quantities, agree with the theory. Owing to the resonant absorption in the potassium vapor, it is difficult to judge from the observed contour whether there is broadening due to the "shock wave" of the pulse envelope. However, it is seen from the obtained spectrograms (see, for example, Fig. 2) that the wing is stretched out predominantly in the long-wave line. There is a wing of appreciable intensity beyond the resonant absorption line.

We have also carried out certain investigations of the time characteristics of the radiation. With the aid of optical delay, we obtained on the same oscillogram pulses before and after the entrance into the cell with the potassium. No noticeable distortion of the pulse waveform was observed, indicating that the main mechanism of broadening is apparently phase modulation.

In Sec. 2 we gave an expression for the group velocity of the propagation of the pulse in a dispersive

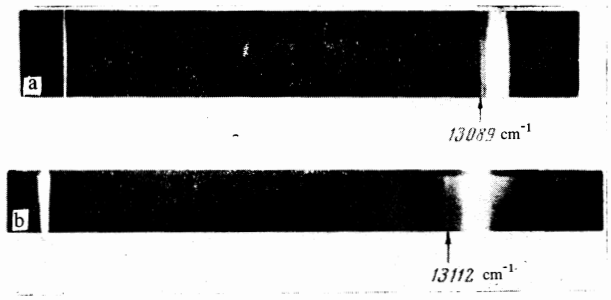

FIG. 4. Emission spectrogram: a – three-photon scattering line, b – five-photon scattering line.

medium; for potassium vapor with $N = 4 \times 10^{16}$, $\epsilon = 2 \times 10^{12}$, $f_{ik} = 0.7$ (f_{ik}—oscillator strength) this expression yields $v \sim 5 \times 10^9$ in the linear case. Over a length of 1 meter, this leads to a pulse delay of ~15 nsec. Experiment revealed no such delay, a fact that apparently must be attributed to the intensity of the incident radiation $I \sim 10^{26}$ photons/cm^2 sec, at which the delay practically vanishes (see formula (6)).

5. MULTIPHOTON SCATTERING

Upon excitation of potassium vapor by the second Stokes SRS component of chloroform, and intense line appeared in the spectrum, at a frequency $2\omega' - \omega_0 = \omega' + \epsilon$ (1309 cm^{-1}) (Fig. 4a). The emission at this frequency can be attributed to the process of three-photon scattering (see Sec. 2), analogous to that observed in[7] in rubidium vapor. Under the conditions of our experiment, the Stark shift $\epsilon\eta$ is small, and therefore amplification is observed at the frequency $2\omega' - \omega_0$.

We note that when potassium vapor is excited by the first Stokes SRS component of nitrobenzene, as was done earlier in[3,5], no three-photon scattering could be observed. This can be attributed to the fact that at low detunings there occurs a strong broadening of the incident radiation and the three-photon scattering does not lead to formation of an individual line.

In this experiment, just as in[3,5,7], intense electron Raman scattering was observed from the $4P_{3/2}$ level, although the line of the three-photon scattering was likewise not observed in this case quite frequently. Whereas upon excitation of potassium vapor by the first Stokes SRS component of nitrobenzene the population of the level (which is needed for the electronic SRS) could be partly attributed to real absorption, in the case of chloroform this is impossible, owing to the large detuning. Therefore, the main mechanism of populating the $4P_{3/2}$ level remains three-photon scattering, and it must be therefore assumed that the practice of three-photon interaction takes place always in those cases when a line of electronic SRS appears on the spectra.

We have also observed intense five-photon emission at the frequency $3\omega' - 2\omega_0 = \omega' + 2\epsilon$ (Fig. 4b) and weaker components with frequencies $\omega' - 2\epsilon$ and $\omega' - 3\epsilon$. The reason for the appearance of these lines may be phase modulation in the presence of the fundamental and three-photon emissions. It must be emphasized that in this case there is frequently no three-photon scattering line.

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