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DISPERSION OF RESONANT NONLINEAR SUSCEPTIBILITY IN POTASSIUM VAPOR

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1. We report in the present article the results of an experimental investigation of the dispersion of the nonlinear susceptibility of potassium vapor near the transitions $4S_{1/2} - 4P_{3/2}$(ν_{01} = 13043 cm^{-1}) and $4S_{1/2} - 4P_{1/2}$(ν_{02} = 12985 cm^{-1}). The use of a source in the form of a powerful pulsed parametric light generator (PLG) with tunable frequency, and observation of self-modulation, self-focusing, and self-defocusing effects have made it possible to trace for the first time the dispersion of the modulus and of the sign of the nonlinear susceptibility in the entire frequency region ($\nu > \nu_{01}$, $\nu_{01} > \nu > \nu_{02}$, $\nu < \nu_{02}$).

New circumstances that distinguish self-action in a vapor from self-action in a condensed medium are the strong influences of saturation of nonlinearity and of the group-velocity dispersion. Both factors became noticeably pronounced in the experiments described below.

2. A PLG based on an $LiIO_3$ crystal was excited by the second harmonic of a one-mode neodymium laser and operated both in the single-resonator and in the two-resonator schemes. The generator pulse duration was $\tau_g = 7 \times 10^{-9}$ sec, the maximum power was 1 MW and the beam diameter ranged from 1 to 2.5 mm. Depending on the operating regime and on the resonator system, the line width ranged from 0.5 to 15 cm^{-1}; the broad spectrum was made up of clearly pronounced mode groups (the so-called "clusters" of the PLG, see Fig. 1).

3. To study the dispersion of the nonlinear susceptibility we investigated the frequency dependence of the self-action effects in potassium near the resnant transitions. Self-action in vapor was observed earlier at fixed frequencies in [1 - 4]; quantitative data on the cubic susceptibility of potassium at $\nu = \nu_{01}$ + 12 cm^{-1} are contained in [2]. We investigated the broadening of the frequency spectrum, self-focusing, and self-defocusing. Figure 1 shows typical spectrograms of the broadened spectra; Fig. 2 shows experimental plots of the relative broadenings $Y = \Delta\nu_{out}/\Delta\nu_{in}$ of the spectrum of the radiation passing through the vapor.

We see that at $\nu - \nu_{01} > 70$ cm^{-1} there is practically no broadening; as ν approaches ν_{01} from the high-frequency side, the broadening first increases and then decreases, vanishing at the resonance. In the region $\nu_{01} > \nu > \nu_{02}$ the experimental $Y(\nu)$ plot has two maxima: Y_{min} corresponds to $\nu \simeq \nu_{02}$ +17 cm^{-1}.

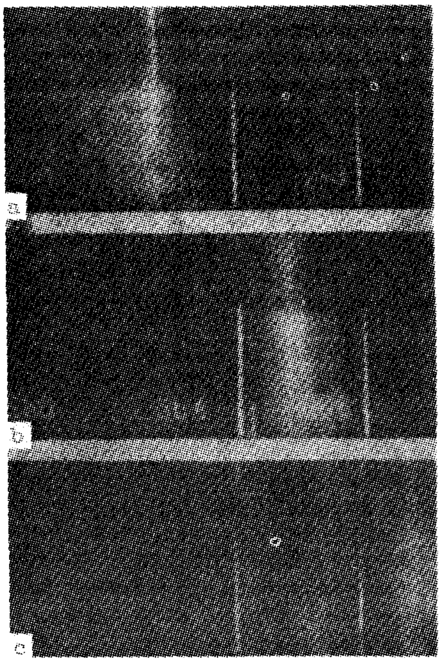

Fig. 1. Typical spectrograms of spectra at the entrance (upper part of spectrogram) and exit (lower part) of a cell with potassium vapor. The spectrogram of Fig. 1a corresponds to a frequency deviation $\nu - \nu_{01} = 51\ cm^{-1}$, power P = 500 kW, and t = 180°C, in Fig. 1b $\nu - \nu_{02} = 34\ cm^{-1}$, P = 1 MW, and t = 180°C, in Fig. 1c $\nu_{02} - \nu = 23\ cm^{-1}$, P = 215 kW, and t = 180°C.

Weaker broadening (at the same beam power) remains at $\nu < \nu_{02}$. The indicated behavior was observed at t = 160 - 280°C; it was impossible to observe nonlinear effects at t > 280°C. The measurements were performed with an unfocused beam; the cell with the vapor had a length ℓ = 35 cm.

Investigations of the near and far field gave the following results. In the region $\nu > \nu_{01}$ we observed clearly self-focusing of the PLG beam; it was revealed by the "hot" points in the near field and by the larger divergence in the far field. The qualitative picture was analogous to that described in [2]. In the regions $\nu_{gr} < \nu < \nu_{01}$ (see Fig. 2) and $\nu < \nu_{02}$, to the contrary, self-defocusing was observed; the beam diameter increased there both in the near and in the far field. The measured values of the beam divergences after passing through the vapor at $\nu < \nu_{02}$ are listed in the table (which gives the values of the divergence angle θ at the entrance and exit of the cell with the vapor, the corresponding values of the beam radius a, and its power P).

4. The nonlinear changes in the angular spectrum (self-focusing, self-defocusing) are satisfactorily described by the results of the theory (see [5, 7]) of the nonlinear refractive index for a two-level system. The Stark effect (see [8]) could be neglected. At frequency deviations $\Delta\nu = \nu - \nu_{0i} \gg T^{-1}$ (where T is transverse relaxation time) and at power not exceeding $10^3 - 10^4$ W, the refractive index of the vapor is

$$n = n_0 + n_2 |E|^2. \qquad (1)$$

Estimates of n_2, obtained from experiments on self-focusing in the region $\nu > \nu_{01}$, are in satisfactory agreement with the data of [2]. The results of experiments on defocusing at $\nu < \nu_{02}$, obtained under conditions when (1) is valid and at moderate powers, were reduced by means of the formula

θ_{in}	θ_{out}	a, mm	P, kW	$\nu_{02} - \nu$	t°, C
0.00086	0,0045	0.5	22.8	79.5	170
–	0,0017	0.8	129.0	4.1	160
–	0,0034	0.7	79.0	19.2	120
–	0,0022	0.7	65.7	16.0	120
–	0,0018	0.5	41.0	31.0	120

$$n_2 = \frac{[\theta_{out} - \theta_{in}] n_o a}{E_o^2 \ell},$$

which is valid for external defocusing. The values of $|n_2|$ determined in this manner (here $n_2 < 0$) at t = 150°C and $\Delta\nu$ = 20 - 30 cm^{-1} are $|n_2| = (1 - 3) \times 10^{-11}$ cgs esu. The calculated values of n_2 (with allowance for the difficulty in determining the vapor concentration) are in satisfactory agreement with the experimental data.

5. The observed spectrum broadenings were naturally interpreted as a result of AM-PM conversion (far from resonance, the shape of the envelope changes little on leaving the vapor; the resonant four-photon parametric luminescence power is negligibly small).

The qualitative form of the Y = Y($\Delta\nu$) curves (see Fig. 2) agrees with the notions concerning the behavior of the nonlinear susceptibility of a two-level system. Worthy of attention is the point between the resonant lines ($\nu_{gr} = \nu_{02} + \tilde{\Delta}$), in which Y passes through a minimum. Its appearance must be ascribed to cancellation of the contributions made to the nonlinear refractive index by the lines ν_{01} and ν_{02}. If we use (1) for n,

$$Y = n_2 k \ell E_o^2, \qquad n_2 = \frac{2\pi p^4}{\hbar^3 \Delta\nu^3}, \tag{2}$$

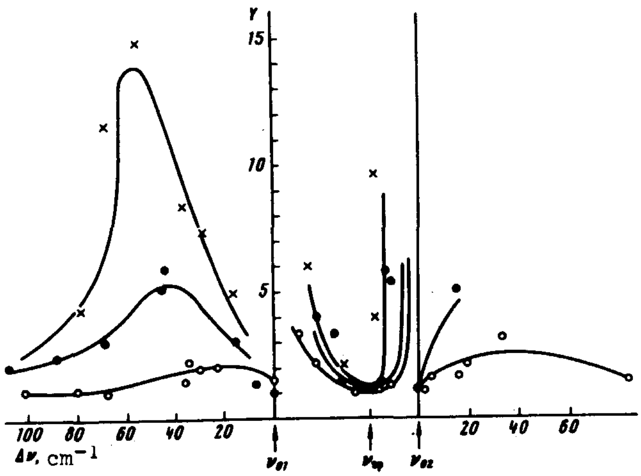

Fig. 2

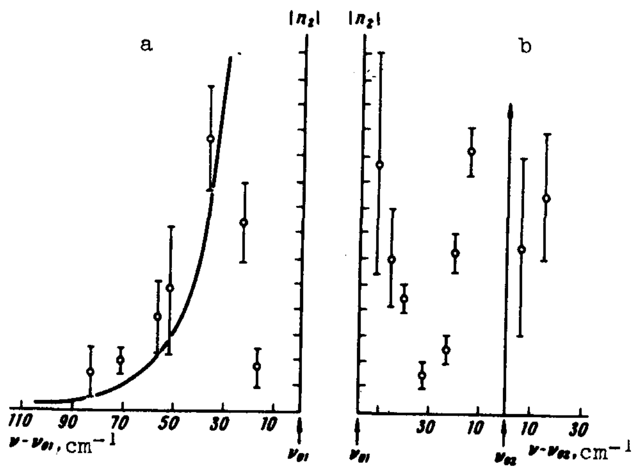

Fig. 3

Fig. 2. Experimental values of relative broadening of the spectrum in potassium vapor as functions of the deviation of the average radiation frequency of the PLG relative to the frequencies ν_{01} and ν_{02}. Vapor temperature T = 200°K. Different curves correspond to different PLG powers (o - intensity I = 5 MW/cm^2, • - I = 16 MW/cm^2, × - I = 26 MW/cm^2).

Fig. 3. Dispersion of $|n_2|$ in the region of the resonant lines of potassium; the region $\nu > \nu_{01}$ (Fig. 3a) was measured at T = 220°C (the solid curve is a plot of the theoretical formula without allowance for saturation). The region $\nu_{01} > \nu$ (Fig. 3b) was measured at t = 200°C. The values of $|n_2|$ are given in relative units.

then we obtain $\tilde{\Delta} = 20\ cm^{-1}$ (see Fig. 2); we note that $\tilde{\Delta}$ (as seen from Fig. 2) changes little within the range of power used by us[1)].

Using (2) we can estimate n_2 from the data on Y (see Fig. 2). In the region of applicability of (1), besides plotting the experimental values of n_2, we constructed a continuous curve in accordance with formula (2) (Fig. 3a). The data obtained for $\nu < \nu_{02}$ agree with the data obtained from defocusing. An important role is played in the resonance regions by saturation effects (including saturation due to the broadened spectrum itself), and the description of the experimental data in terms of n_2 is arbitrary. We note that in the entire region $\nu > \nu_{01}$ the experimental values of n_2 are higher by approximately one order of magnitude than the theoretical ones, owing to the increase of the field intensity as a result of self-focusing.

6. The structure of the broadened spectra differs from that known for liquids (if the absorption is negligible, the spectra are symmetrical but the phase relations are disturbed). Estimates show this to be connected with the influence of the group-velocity dispersion (this was indicated in [6] as applied to experiments with vapor).

In the resonance region, under our conditions, nonstationary coherent effects are possible, wherein the period of the amplitude modulation of the PLG radiation is comparable with the characteristic relaxation times T_1 and T_2 of the resonant transition.

7. The considerable resonant nonlinearity permits realization of an effective tunable four-photon laser and also third-harmonic generation under certain conditions. At intense narrow-band pumping, we observe narrow lines that can be ascribed to four-photon generation.

8. We note in conclusion that the use of a PLG in nonlinear spectroscopy uncovers also other possibilities not realized in the present study. The PLG is of necessity a multifrequency generator; in our scheme there are time-synchronized giant pulses at the frequencies ν_1, ν_2, $\nu_n = \nu_1 + \nu_2$, and $\nu_n/2$. This can be used for combination effects, for probing-signal schemes to register polarization or level-population difference, etc.

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