Self-focusing in potassium vapor under two-photon resonant excitation

S. A. Bakhramov, U. G. Gulyamov, K. N. Drabovich, and Ya. Z. Faĭzullaev

Nuclear Physics Institute, Uzbek Academy of Sciences
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Self-focusing of laser radiation was observed for the first time in two-photon resonance with 4*s*–4*d* 4*s*–6*s* transitions in potassium vapor. The critical self-focusing intensities were measured. The experimental data agree well with the theoretical ones.

1. We observed, for the first time, self-focusing of laser radiation in two-photon absorption in potassium vapor. A distinguishing feature of self-focusing in two-photon resonance is the nonlinear dependence of the refractive index on the square of the intensity of the incident radiation. The significant parameter in this case is not the critical power but the critical intensity of the self-focusing of the laser beam. We measured the critical intensities of the self-focusing in the case of two-photon resonance. Good agreement was obtained between the theoretical and experimental data.

2. Self-focusing in single-photon resonance was first observed in[1] and was subsequently studied in detail. The possibility of self-focusing in two-photon resonance was indicated in[2] (see also[3]), but this effect was not observed experimentally to this day. The study of self-action of laser radiation at two-photon resonance is of particular interest at present in view of the extensive use of two-photon resonance in nonlinear optics, both for frequency conversion (see, e.g.,[4]) and for ultrahigh-resolution spectroscopy.[5]

3. To observe self-focusing under two-photon resonance conditions, we used a dye laser excited by a single-pulse ruby laser. Smooth tuning of the lasing frequency with the aid of a diffraction grating in the range 7150–7400 Å has made it possible to pass in succession through the two-photon resonance with the 4*s*-6*s* and 4*s*-4*d* transitions in potassium vapor. The

(a) (b)

FIG. 1. Distribution of the intensity in the incident beam (a) and in the beam after passing through the potassium vapor (b).

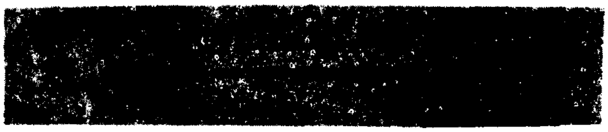

FIG. 2. Side-view photograph of the cell in the presence of self-focusing.

Transitions	T° C	I_{cr}^{exp}, MW/cm²	I_{cr}^{theor}, MW/cm²
4s-6s	280	16.2	24
	320	9.2	14
4s-4d	280	90	55
	320	50	39

dye-laser pulse duration τ_p amounted to 35 nsec at a maximum power ~800 kW. The spectral width of the generation line was $\gamma \approx 2$ Å, and the beam divergence was ≈15′. The dye-laser beam was focused by a lens of focal length 40 cm onto the entrance window of a cell with potassium vapor ($l = 30$ cm), so that the beam passing through the cell was known to diverge. The spectral composition of the incident and transmitted radiation was investigated with a DFS-8 spectrograph and an SPM-2 monochromator with infrared receiver. The transverse distribution of the intensity in the incident and transmitted beams was photographed with cameras placed 80 cm away from the focusing lens. The potassium vapor pressure was varied between 0.001 and 1.2 mm Hg. Two-photon excitation of the potassium vapor was accompanied by luminescence in the visible region, so that the spatial characteristics of the beam could be investigated by photographing the cell from the side.

The following experimental results were obtained. When the doubled generation frequency coincided with the frequencies of the 4s-6s and 4s-4d transitions, an abrupt decrease of the divergence of the transmitted radiation was observed, accompanied by an increase of the intensity at the center of the beam (Fig. 1). Figure 2 shows a photograph of the beam taken from the side of the cell and illustrates the spatial distribution of the luminescence of the potassium vapor under two-photon excitation.

The self-focusing effect disappeared when the deviation from the two-photon resonance with the 4s-4d and 4s-6s transitions amounted to 2—3 Å in either direction.

We measured the threshold intensities of the self-focusing and investigated their dependence on the potassium-vapor temperature (see the table). Investigation of the spectra of the incident and transmitted radiation revealed no noticeable change in the spectral width of the laser radiation even under the self-focusing conditions.

4. Theoretical calculations using the known equations for two-photon resonance[3] show that at the considered transitions the principal contribution to the field-induced change of the refractive index is made by the difference between the nonresonant polarizabilities κ_i of the levels 4s and 6s or 4d. Consequently, we can confine ourselves in the estimates to a calculation of the population difference q between the ground and excited states, averaged over the random factors of the field. Recognizing that in our case $\tau_p \ll T_1$ or T_2 (T_1 and T_2 are the longitudinal and transverse relaxation times), and that $\gamma \gg 1/\tau_p$, we obtain

$$\bar{q} = q_0 \exp\left[-\frac{8r^2\gamma}{4\gamma^2+\delta^2}\int_{-\infty}^{t} I^2(t')dt'\right] \quad \text{at } |\delta| < \gamma, \tag{1}$$

where q_0 is the initial population difference, τ is the composite matrix element of the two-photon transition,[3] $\delta = \omega_{21} - 2\omega$ is the deviation of the frequency ω of the exciting field from two-photon resonance with the corresponding transition, and I is the intensity of the exciting field.

Expanding (1) in a series and replacing the integral by the approximate value $I_0\tau_p$ (I_0 is the peak intensity), we obtain after substitution in the expression for the refractive index under two-photon resonance[3]

$$n = n_0 + n_4 I_0^2, \qquad n_4 = 8\pi N(\kappa_2 - \kappa_1) q_0 \frac{r^2\gamma\tau_p}{4\gamma^2+\delta^2}, \tag{2}$$

where N is the density of the particles. For the 4s-6s and 4s-4d transitions in potassium vapor we have $\kappa_2 - \kappa_1 \approx 3.7\times10^{-22}$ and 4.6×10^{-22}, $\tau \approx 0.8\times10^{-5}$ and 0.13×10^{5} (cgs esu), respectively.[1)] The calculated data for the critical intensity I_{cr} are given in the table.

The difference between the experimental and theoretical values is due both to the approximations used in the calculation of the refractive index and to the approximation of the multimode beam by a Gaussian beam.

It should be noted that a definite contribution can be made to the nonlinear dependence of the refractive index on the field by single-photon resonance with the transition 4s-4p. Estimates show, however, that the critical intensity of the self-focusing in the interaction of radiation with only the indicated transition in the 7400 Å region is 10^{-3} times the values listed in the table. The insignificance of this contribution is confirmed also by the fact that in the case of detuning from the two-photon resonances in the low-frequency direction, and consequently when the resonance with the transition 4s-4p is approached, the self-focusing vanishes in full accord with formulas (1) and (2).

5. Just as in the case of two-photon stepwise excitation of potassium atoms (see, e.g., [7]), we have observed in our experiments directional radiation in the violet and infrared regions, corresponding to the transitions from the 4d and 6s levels. Under the self-focusing

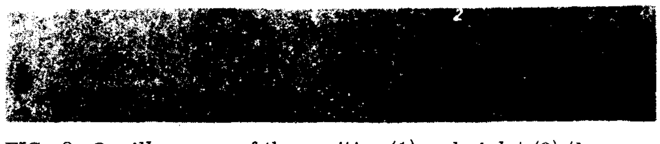

FIG. 3. Oscillograms of the exciting (1) and violet (2) (λ = 4047 Å) pulses in the presence of self-focusing.

conditions, the intensity of the violet radiation increased significantly, so that we were able to determine its time dependence. Figure 3 shows oscillograms of the exciting (1) and of the violet (2) ($\lambda = 4047$ Å) pulses. We see that the violet pulse is approximately 1.7 times shorter than the exciting pulse.

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[1)]To calculate $k_2 - k_1$ and τ we used the dipole-moment values given in[6].

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