Vavilov-Cherenkov effect under conditions of near-resonant interaction of intense light beams with atomic potassium vapor

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A model is suggested for a six-photon parametric process and explains a number of features observed in the frequency range of the main doublet in the spectra of light scattered in a cell containing potassium vapor. Under strong dispersion conditions a nonlinear polarization induced in the medium and associated with the parametric process may travel at a velocity exceeding that of light, giving rise to a situation typical of the Vavilov-Cherenkov effect. Experimentally obtained spectrograms demonstrate the influence of the processes which are explained by the proposed model.

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1. INTRODUCTION

Cherenkov radiation emitted by a bundle of electromagnetic rays traveling in a medium[1] may result from a non-linear interaction between two or three coherent light waves.[2-4] In this case the source of Cherenkov radiation is a nonlinear polarization traveling in the medium at a velocity exceeding that of light (superluminal velocity). If such polarization is associated with difference frequencies,[3] the effect can be used to generate coherent radiation in the far infrared. However, observation and study of this effect meet with certain difficulties. More favorable conditions for the observation of Cherenkov radiation due to a nonlinear polarization propagating in a medium is provided by a situation of four-photon interaction associated with stimulated Raman scattering. According to the results of Ref. 4, the vertex angle of the output radiation cone obtained in experiments on Raman stimulated scattering can be calculated from the condition of emission of Cherenkov radiation

$$\cos\theta = k_i/k, \qquad (1)$$

where k and k_i are the wave numbers of a diverging light wave and a wave of nonlinear polarization induced in a medium.

The application of this approach in the present case meets with difficulties of interpretation of a number of specific features of the frequency and angular spectra of the output radiation obtained after passage, through a cell with atomic potassium vapor, of intense coherent radiation whose frequency at the entry to the cell corresponds to the region occupied by the main doublet $4S_{1/2}$–$4P_{3/2}$ ($\nu_{01} = 13\ 046\ \text{cm}^{-1}$) and $4S_{1/2}$–$4P_{1/2}$ ($\nu_{02} = 12\ 989\ \text{cm}^{-1}$). The absence of symmetry in the frequency-angular spectrum of the output radiation limits[5] the validity of the model of four-photon parametric scattering used in Refs. 5 and 6 to interpret the experimental results. It seems to us that the application of a model of six-photon parametric scattering[7-9] may overcome these difficulties. The compatibility of the conditions of vector phase matching, which follow from the four-photon parametric scattering model, with the characteristics of the angular structure of the output radiation spectrum ($\nu < \nu_{01}$ case) allows us to consider the phenomenon under study as the Vavilov–Cherenkov effect.

2. STIMULATED RAMAN AND PARAMETRIC SCATTERING IN A TWO-LEVEL MEDIUM

The starting point of the model used in the present study is that in the case of a resonant interaction of optical radiation with atomic potassium each of the doublet ($4S_{1/2}$–$4P_{1/2}$, $4S_{1/2}$–$4P_{3/2}$) transitions is characterized by final states of the opposite parity. Since these transitions are of the electric-dipole allowed type, it follows that three-photon processes are next in importance[10,11] after one-photon absorption, spontaneous emission, and stimulated emission. A suitable example is provided by three-photon electron Raman scattering (Fig. 1a), whose stimulated analog was predicted for the optical range in Ref. 12 and the first observations were reported in Ref. 13.

Interaction between an intense coherent pump wave of frequency ν (case $\nu < \nu_{01}$, $\nu_{01} - \nu \ll \nu_{01}, \nu_{01}$, where ν_{01} is the frequency of the $4S_{1/2} - 4P_{3/2}$ transition) with a wave of three-photon electron stimulated Raman scattering, whose frequency[14]

$$\nu_s = 2\nu - \nu_{01} \qquad (2)$$

is in this case shifted to the Stokes region (relative to the pump frequency), is responsible for the appearance of a nonlinear polarization in the medium and the propagation of such polarization is related to the emission of Cherenkov radiation. This interaction presupposes

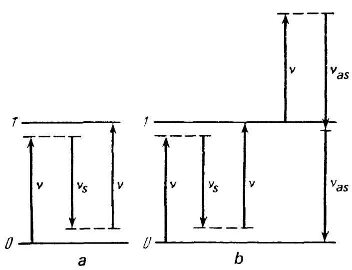

FIG. 1. a) Elementary processes of three-photon scattering of light.[14] b) Elementary processes in the case of six-photon parametric scattering.

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the existence of a resonant parametric process with six photons participating in each elementary event (Fig. 1b):

$$3\nu=\nu_s+\nu_{as}'+\nu_{as}''. \quad (3)$$

In the degenerate case, usually characterized by the maximum rate of growth of parametric waves,[15] we have

$$\nu_{as}'=\nu_{as}''=\nu_{as}, \quad (4)$$

where

$$\nu_{as}=(\nu_{01}+\nu)/2. \quad (5)$$

Growth of the waves associated with this resonant six-photon process may overtake growth of the waves associated with lower-order nonlinear processes.[8] If a medium has properties which ensure satisfaction of the phase matching conditions, which in this case ($\nu < \nu_{01}$) have the form

$$3\mathbf{k}=\mathbf{k}_s+\mathbf{k}_{as}'+\mathbf{k}_{as}'', \quad k_{as}'=k_{as}'', \quad (6)$$

the above circumstances should make it possible to observe the radiation associated with this process.

The simplified equations for the amplitudes of the Stokes A_s and anti-Stokes A_{as} waves, allowing for the Raman and parametric processes, have the following form in the approximation of a constant pump field[8,15] (steady-state case):

$$\partial A_s/\partial z=r_1I^2A_s-r_2A^3A_{as}^*A_{as}^* \times\exp[-i(3\mathbf{k}-2\mathbf{k}_{as}-\mathbf{k}_s)z], \quad (7)$$

$$\partial A_{as}/\partial z=r_3II_{as}A_{as}+r_4A^3A_s^*A_{as} \times\exp[-i(3\mathbf{k}-2\mathbf{k}_{as}-\mathbf{k}_s)z], \quad (8)$$

where r_1, r_2, r_3, r_4 are the nonlinear coupling coefficients; I and I_{as} are the intensities of the waves involved.

Equations (7) and (8) are simplified by dropping the terms representing the four-photon interactions of the $\nu+\nu_s$ and $\nu+\nu_{as}$ type which generally can limit the growth of the waves participating in the six-photon process, but the very existence of these four-photon processes[16] is possible only after the appearance of initiating radiation at frequencies ν_s and ν_{as}.

The diagram (Fig. 2) corresponding to the vector phase matching conditions (6) can be used to calculate the vertex angle of the cone along whose generators we can expect (because of the axial symmetry) growth of the radiation of frequency ν_{as} This angle can be found from

$$\theta=\arccos\frac{3k-k_s}{2k_{as}}. \quad (9)$$

If the wave number of the induced nonlinear polarization vector

$$k_i={}^3/_2k-{}^1/_2k_s, \quad (10)$$

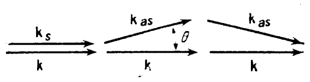

FIG. 2. Vector diagram for calculating the angles of scattering of anti-Stokes components ν_{as} ($\nu < \nu_{01}$ case).

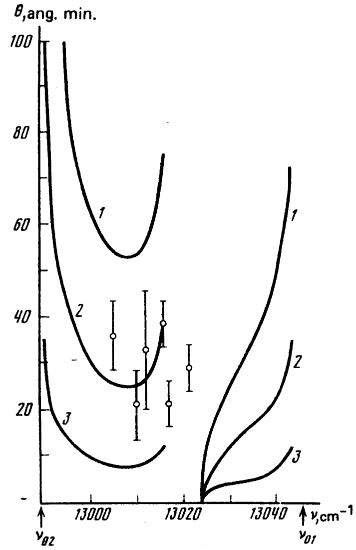

FIG. 3. Dependences of the scattering angle of the anti-Stokes radiation on the pump frequency calculated from Eq. (9): variation of the temperature of the potassium vapor altered the expected value of the scattering angle. The calculations were carried out for the following potassium vapor temperatures t (°C): 1) 280; 2) 230; 3) 180. The experimental results (○) were obtained at t = 200–220 °C.

governed by the combination of the pump vector and the vector representing the three-photon electron stimulated Raman scattering, is less than the wave number of the vector of the radiation emitted at a frequency ν_{as}, it follows from Eq. (1) that this polarization can act as a source of Cherenkov radiation.

The existence in the atomic potassium vapor, in the frequency range of the main doublet, of parts of the spectrum satisfying the inequality

$$3k<k_s+2k_{as}, \quad (11)$$

leads us to expect—in accordance with Eq. (9)—a dependence of the Cherenkov angle of the scattered radiation at the frequency ν_{as} on the pump frequency. The results of the relevant calculations are presented in Fig. 3 for a set of temperatures. It is clear from this figure that the dependence of the scattering angle on the pump frequency calculated on the basis of Eq. (9) has a discontinuity due to the fact that the condition (11) is not satisfied at frequencies 13 018-13 025 cm^{-1}. This discontinuity is due to the $4S_{1/2}$–$4P_{1/2}$ transition.

3. DESCRIPTION OF APPARATUS

The interaction of coherent radiation with atomic potassium vapor was studied using apparatus shown schematically in Fig. 4. The source of tunable radiation was an optical parametric oscillator (OPO) excited by the second harmonic of a glass neodymium laser. The output power of the OPO was 100-500 kW in the form of 15 nsec pulses. The structure of the frequency spectrum of the output radiation characteristic of the OPO consisted of 1–3 separate lines, which were separated by not more than 3 cm^{-1}. The width of each line was 0.2 cm^{-1}.

The output frequency was tuned by rotating a nonlin-

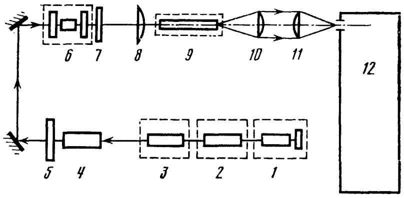

FIG. 4. Schematic diagram of the apparatus: 1)–3) laser oscillator with a glass element activated with neodymium ions, and a system of amplifiers; 4) frequency doubler (KDP crystal); 5) SZS-21 filter; 6) optical parametric oscillator; 7) KS-18 filter; 8) lens (f = 20 cm); 9) cell with potassium vapor; 10), 11) system of lenses imagining the exit window of the cell onto the spectrograph slit; 12) DFS-8 spectrograph.

ear crystal ($LiIO_3$) in the OPO resonator. The divergence of the light beam emerging from the OPO did not exceed 0.0009 rad and the beam diameter was 1–3 mm. A weakly focusing lens (f = 20 cm) placed at the entry to the potassium vapor cell enhanced the intensity of the exciting radiation.

The distance between the lens and the potassium vapor was selected so that the center of the cell coincided with the narrowest part (constriction) of the beam. In this case the diameter of the beam at the exit from the cell did not exceed 2 mm when the vapor was at room temperature. An optical system of lenses 10 and 11 projected, on the 1:1 scale, the structure of the radiation field from the plane of the exit window of the cell onto the slit of a DFS-8 spectrograph (dispersion 6 Å/mm).

This method made it possible to record spectrograms of the scattered radiation and at the same time determine the distance (relative to the axis of the pump beam) of those points on the exit window of the cell whose positions were governed by the frequency components of the scattered radiation emerging at an angle from the potassium vapor cell. The spectrograms obtained under these conditions could be used to calculate the angles of divergence of the various frequency components of the output radiation.

4. EXPERIMENTAL RESULTS. DISCUSSION

Propagation of an intense coherent radiation beam across atomic potassium vapor near the $4S_{1/2}$–$4P_{1/2}$, $4S_{1/2}$–$4P_{3/2}$ doublet may be accompanied by a number of nonlinear-optics effects such as self-focusing,[17,18] two- and three-photon electron stimulated Raman scattering,[12,13,19] and four-photon parametric scattering.[16]

The effect considered in the present study ($\nu < \nu_{01}$ case) is characterized by at least two features which can be used to separate this effect from the others. Firstly, we can expect a change in the frequency spectrum of the output radiation resulting in asymmetry which should correspond—according to Eqs. (2) and (5)—to the condition

$$2(\nu_{as}-\nu)=\nu-\nu_s. \qquad (12)$$

Secondly, the output radiation should contain components with scattering angles obeying the condition (9). The strong dispersion of the medium near the resonance, possibility of changes in this dispersion with temperature, and the ability to tune the frequency of the exciting radiation all facilitate the identification, in the output spectrum from the cell, of the features associated with the effect under study.

Figure 5a shows a spectrogram recording the frequency and spatial structure of the output radiation. The reference arrows in the spectrogram identify the positions of the investigated doublet lines. The pump frequency obeys $\nu < \nu_{01}$, where ν_{01} is the frequency of the $4S_{1/2}$–$4P_{3/2}$ transition. The exciting radiation frequency generated with the OPO consists of two narrow-band components. This factor and also the influence of the Stark effect[20] transform the radiation corresponding to the frequency ν_s of three-photon electron stimulated Raman scattering into a broad line whose structure produces only the general features of the pump spectrum. In the anti-Stokes (relative to the pumped) region the spectrum of the output radiation is limited by the frequency ν_{as}, which is given by Eq. (5). On the whole, the frequency spectrum of the output radiation is asymmetric in accordance with the condition (12) of the proposed model.

The presence in the output radiation spectrum, near the pump frequency, of a diffuse background is clearly associated with the influence of the four-photon parametric scattering processes[16] and of the amplitude-phase modulation.[18,21]

Since in the process of recording a spectrogram the whole exit window of the cell is projected onto the spectrograph slit (Fig. 5b), only part of the radiation corresponding to the vertical slit, 0.22 mm wide, enters the spectrograph so that the spectrogram reflects

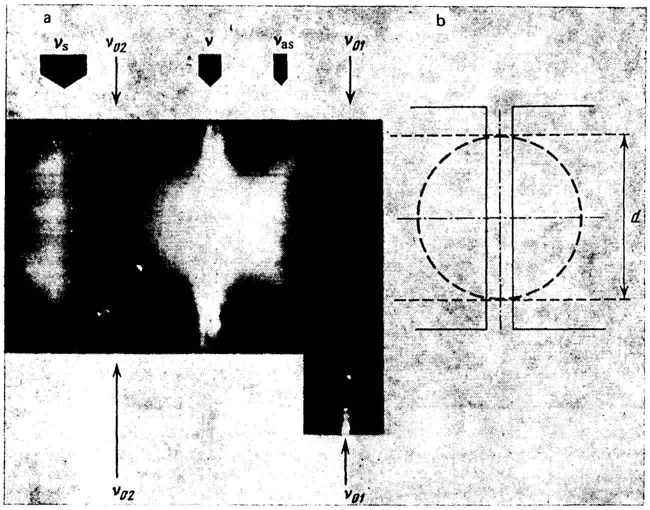

FIG. 5. a) Scattered-radiation spectrum ($\nu_{01} - \nu \approx 30$ cm^{-1}). The energy of the pump pulses at the entry to the cell was $P \approx 3$ mJ and the duration of the pulses was $\tau = 15\times10^{-9}$ sec. The arrows labeled ν_s and ν_{as} identify the frequencies given by Eqs. (2) and (5); ν_{01} and ν_{02} are the potassium reference lines. b) Image of the exit window of the cell super-imposed on the spectrograph slit. The dashed circle represents the intersection of a conical surface, formed by the radiation scattered in the cell and having the frequency ν_{as}, with the plane of the exit window of the cell.

the distribution on the exit window of the cell of those frequency components which travel in the vertical plane along directions other than the direction of the pump wave.

It is clear from the spectrogram that in the part of the spectrum corresponding to the frequency ν_{as} there are indeed frequency components propagating at an angle to the optic axis coinciding with the direction of propagation of the pump wave. Clearly, because of the axial symmetry and isotropy of the medium these frequency components travel along generators of a cone whose axis coincides with that of the optic axis.

Spectrograms obtained earlier[5,6] under similar experimental conditions ($\nu < \nu_{01}$) confirm the existence of a diverging cone of scattered radiation whose frequency is shifted in the anti-Stokes direction relative to the pump wave.

If we know the length of the potassium vapor cell (300 mm) and the distance d (Fig. 5), we can estimate the angle of divergence of the components of the scattered radiation in the range of the frequency ν_{as}. Such estimates were made for some values of the exciting frequency. The angular divergence of these components (Fig. 3) agreed with the divergence calculated from Eq. (9) on the basis of the above model.

The temperature of the potassium vapor in the cell was 200–220 °C. In the selected part of the spectrum the influence of self-interactions[17,18] hindering the separation of the investigated effect was minimal.[18]

The agreement between the proposed model and our experimental data, and the ability to explain the experimental results obtained by other authors,[5,6] allow us to assume that the process of six-photon parametric scattering occurs in the investigated spectral range in potassium vapor excited by intense coherent radiation. This process can be regarded as the Vavilov–Cherenkov effect.

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