Instability of a laser beam with a negative detuning in a resonant medium

D. V. Gaĭdarenko, A. G. Leonov, A. A. Panteleev, V. A. Roslyakov,
A. N. Starostin, and D. I. Chekhov
Moscow Physicotechnical Institute, 111700, Dolgoprudnyĭ, Moscow Oblast

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The instability of a laser beam with a negative detuning ($\Delta\omega = \omega_L - \omega_0 < 0$) in a resonant medium has been studied experimentally. The beam intensity ranged up to 10^7 W/cm^2; the resonant medium was sodium vapor with a density of 10^{14}–10^{16} cm^{-3}; and the detuning was with respect to the D_1 line of Na. Observations reveal that the scattering contains cone radiation, that scattering components with a frequency shifted from the laser frequency are generated, and that filaments form. This picture of the scattering is similar in many regards to the scattering in the case $\Delta\omega > 0$, which has been studied thoroughly in previous work. The results are compared with theoretical predictions.

Intense laser light propagating through an optically dense resonant medium is unstable because of four-wave mixing. This instability is well known and has been studied in many places (e.g., Refs. 1–6). The onset of this instability is accompanied by an increase in the divergence of laser beam, a broadening of its spectrum, the generation of cone radiation, the generation of several scattering components whose frequency is shifted from that of the laser light (ω_L), etc. In general, all these effects have been observed in the case of a positive detuning of ω_L with respect to the resonant frequency ω_0, since it has been assumed that the degenerate self-focusing,[7] which leads to the breakup of the laser beam into distinct filaments, would raise the interaction intensity substantially in the case $\omega_L > \omega_0$, while at $\omega_L < \omega_0$ the decrease in the power density in the beam due to self-defocusing would make the onset of an instability improbable.

It was shown theoretically in Ref. 8, where four-wave mixing was taken into account, that an intense laser wave undergoes a transverse instability, regardless of the sign of the detuning from resonance, $\Delta\omega = \omega_L - \omega_0$. The development of spatial and frequency instabilities should be thought of as a common process, which leads to a fundamentally new, spectrally nondegenerate instability of a laser beam. A characteristic feature of this instability is an interrelationship between the cone radiation and a small-scale self-focusing. At negative detunings, $\Delta\omega < 0$, the scattering of the laser light observed at the exit from the resonant medium should correspond in a qualitative way to the case $\Delta\omega > 0$. In this letter we report an attempt to experimentally study the interaction between a laser beam and a resonant medium under the condition $\Delta\omega < 0$, with the goal of observing this instability.

The experimental stand consisted of a tunable pulsed dye laser, a heated cell filled with sodium vapor, and diagnostic systems. The energy of the laser beam in the experiments ranged up to 6 mJ; the pulse length at half-maximum was $\tau_{1/2} = 18$ ns. The laser produced a linearly polarized Gaussian beam with a spectral half-width of 0.008 nm, a divergence ~1 mrad, and a diameter of 1.5 mm at the center of the cold cell. The density of the Na vapor in the cell, filled with a buffer gas (argon at a pressure of 10 Torr), was measured by the Rozhdestvenskiĭ hook method. It was also determined from the temperature distribution along the cell wall and a curve of the saturation vapor pressure. The results found by the two methods agreed within ~30%. The length of the column of sodium vapor was $L \simeq 10$ cm. The intensity distribution over the cross section of the laser beam scattered in the vapor was measured with a line array of photodiodes and was photographed. The scattering spectra were recorded with a DFS-452 spectrograph. A slit was positioned in the focal plane of the spectrograph; an FEU-84 photomultiplier was placed behind the slit. The photomultiplier output signal was fed to a V9-5 strobed voltmeter, which measured the amplitude of this signal at the time at which the generation pulse reached its maximum, with a resolution of 4 ns (during which time the intensity of the light remained essentially constant). This signal then went to an IBM PC/AT computer for further analysis. A DFS-452 diffraction grating was rotated by a stepping motor to put various regions of the spectrum on the slit in succession. With the laser generating periodic pulses, an instantaneous spectrum of the light scattered in the vapor was plotted on the computer screen with a resolution of 0.02 nm.

All the experiments were carried out at Na vapor densities $N \simeq 10^{14}$–10^{16} cm^{-3}; the laser intensity J ranged up to 10 MW/cm^2. The detuning in the long-wavelength direction from the sodium D_1 line (which corresponds to the $S_{1/2}$–$P_{1/2}$ transition, with $\lambda = 589.59$ nm) was $\Delta\lambda \simeq 0$–0.3 nm.

The results found here indicate a well-developed instability of the laser beam in

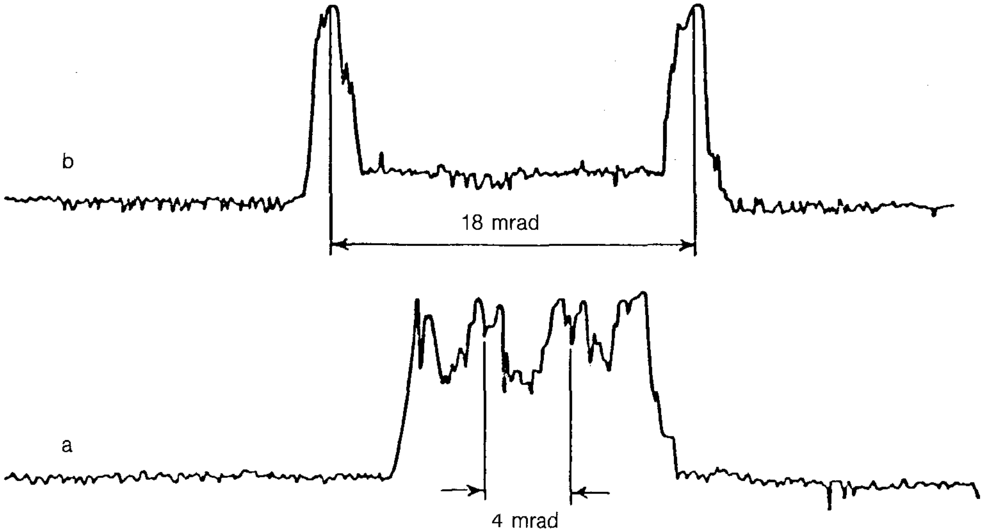

FIG. 1. Intensity distribution along a diameter of the scattered beam at a detuning of 0.21 nm. a—$J = 0.5$ MW/cm^2, $N = 6 \times 10^{14}$ cm^{-3}; b—$J = 6$ MW/cm^2, $N = 6 \times 10^{15}$ cm^{-3}.

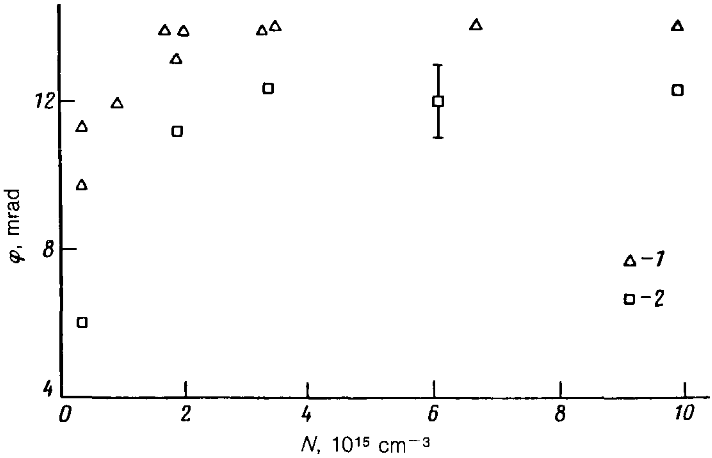

FIG. 2. Angular diameter of the inner ring versus the sodium vapor density. The detuning is 0.21 nm. 1—$J = 3.6$ MW/cm^2; 2—$J = 1.2$ MW/cm^2.

these ranges of values of the interaction parameters. At small values of N and J, two rings appear in the intensity distribution along a diameter of the light beam scattered in the vapor (Fig. 1a). With increasing N and J, the contrast of the outer ring decreases; this ring spreads out and eventually disappears. At this point, only the inner ring is detected in the beam cross section (Fig. 1b). Later, this inner ring also becomes less sharp (previously,[4] in the case $\Delta\omega < 0$, only a single ring has been observed). The angular diameter of these rings, φ, increases with increasing intensity and with decreasing $\Delta\lambda$. With increasing vapor density, φ also begins to increase; it then reaches saturation (Fig. 2). This behavior is quite different from the $\varphi \propto N^{0.4-0.5}$ which has been discussed in the literature.[1-3] We might add that some speckle is detected on photographs of the scattered beam; this speckle corresponds to a breakup of the beam into distinct filaments. In contrast with Refs. 2 and 4, the scattering spectrum has some clearly defined components with a slight shift (Fig. 3). Their frequencies, ω_i (except that of the line at ω'), can be described satisfactorily by a splitting of a three-level system in a strong field.[9] The lines themselves are broadened to 0.05–0.15 nm (the line at ω_{22}^- arises as a component of the strong ω_2^- line). In contrast with the $\Delta\omega > 0$ case, the spectra of both the rings and the light inside them are essentially identical at all observation angles θ. However, outside the ring, at large angles, $\theta \simeq 10$–25 mrad, the intensity of the shifted components, which is essentially independent of θ, reaches a value on the order of the intensity of the line at the laser frequency, which decreases rapidly in this angular region. The frequencies ω_i themselves are almost independent of θ.

Some similar experiments which we carried out with positive detunings reproduced the data of Refs. 1–6.

It was shown in Ref. 8 that the pole of the retarded Green's functions for photons propagating in the presence of an intense laser field is determined by the following

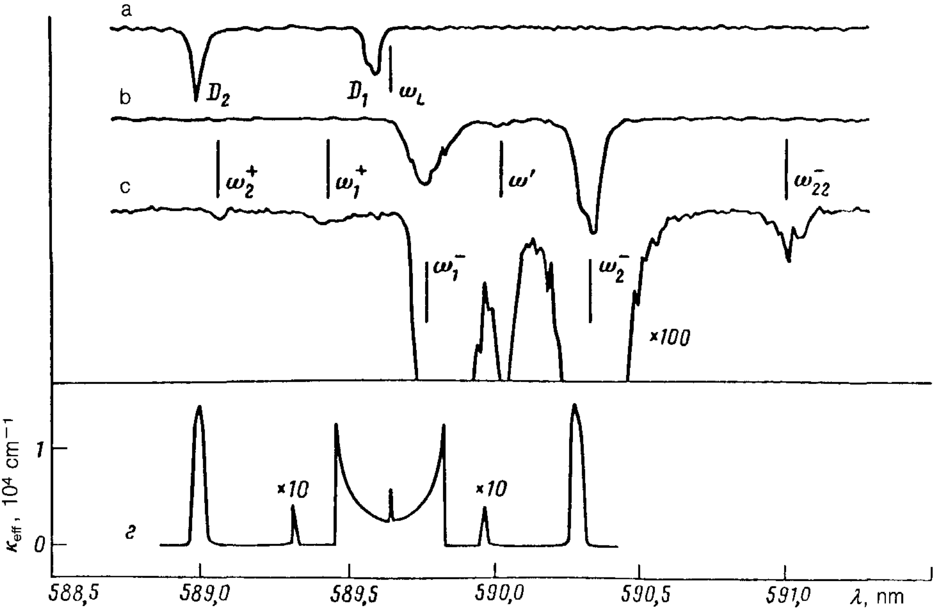

FIG. 3. a: Spectrum of reference D lines of sodium. b,c: Spectrum of the scattered beam at a detuning of 0.05 nm, measured under different conditions. Here $J = 6$ MW/cm^2 and $N = 6\times10^{15}$ cm^{-3}. d: Spectrum of the effective growth rate, calculated for the same conditions.

equation, when four-wave mixing is taken into account:

$$(c^2\vec{k}_1^2 - \epsilon_1\omega_1^2)(c^2\vec{k}_2^2 - \epsilon_2\omega_2^2) - 16\pi^2\omega_1\omega_2\beta_1\beta_2 = 0. \tag{1}$$

Here $\vec{k}_j$ is the wave vector of the photons with frequency ω_j; the wave matching condition $\vec{k}_1 + \vec{k}_2 = 2\vec{k}_L$ holds; $\omega_1 + \omega_2 = 2\omega_L$; and $\varepsilon_j = 1 + 4\pi\alpha_j$. Dispersion relation (1) also follows from the semiclassical description.[3,10] The coefficients α and β are proportional to the corresponding polarization operators from Ref. 8. They describe the polarizability of a test signal in the presence of a laser wave and a four-wave coupling of modes. In contrast with Refs. 3 and 10, we calculated α_j and β_j for a three-level atomic subsystem, since the Rabi frequency of the pump wave is close to the value of the fine-structure splitting of the sodium doublet. The explicit dependence of the relaxation constants on the density of the atomic vapor was taken into account.[11]

Analysis of Eq. (1) shows that the growth rate $\kappa(\nu,\theta) = \mathrm{Im}(\vec{k}_1)$ is a strong function of the angle (θ, between $\vec{k}_1$ and $\vec{k}_L$) and the frequency (the detuning from the laser wave, $\nu = \omega_1 - \omega_L$). Since the wave amplification is determined by the optical length κl, since the aperture of the laser wave is bounded, and since the relation $l(\theta) = d/\sin\theta$ holds, where d is the beam diameter, it is natural to analyze an effective growth rate $\kappa_{\mathrm{eff}} = \kappa/\sin(\theta)$. Figure 3d shows κ_{eff} versus the frequency at small angles ($\theta = 2$ mrad) for the experimental conditions of Fig. 3, b and c. We see that the light at the output should have a spectrally broad composition, in contrast with the spec-

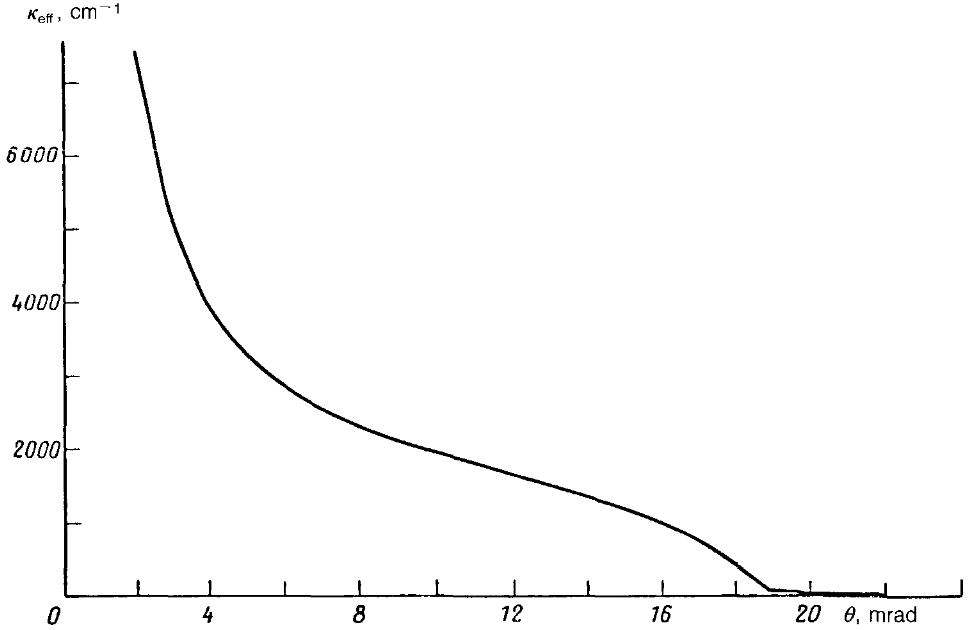

FIG. 4. κ_{eff} versus the angle θ for the conditions of Fig. 3, b and c, for a detuning of the scattered mode from the laser line by 0.17 nm.

trally narrow lines which follow from an analysis of κ. The detunings for these lines, at an intensity $J = 6$ MW/cm^2, are 0.19, 0.57, and 0.76 nm. They correspond to the Rabi splitting of a three-level system.[9] A study of κ_{eff}, on the other hand, shows that a significant amplification occurs in the lines at 0.57 and 0.76 nm only at large angles, $\theta > 100$ mrad. At small angles, only two lines are observed. Their peaks are at detunings of 0.19 and 0.67 nm, in good agreement with the experimental data. There is an unexpected result here: Both the experiments and the calculations reveal the appearance of a slight gain near the frequency ω'; this gain is uncorrelated with the quasienergy spectrum of the three-level system. Analysis of κ_{eff} as a function of θ shows that this quantity has a broad angular distribution (Fig. 4) and retains its shape for a spectrally broad frequency range. These results agrees well with experimental data on the spectral-angular characteristics of the cone radiation. Our calculations of the dependence of the optimum angle θ on the vapor density for this mode yield results in qualitative agreement with the experimental data. These results differ from the $N^{0.5}$ dependence which has been discussed theoretically in the literature.[2,3]

It was found experimentally that a significant fraction of the energy of the laser wave is converted into side modes (Fig. 3b). Further evidence for this conversion comes from the large values of the calculated growth rates (Fig. 3d). The meaning here is that the explanation which has been proposed is valid only in a qualitative sense. In particular, it does not describe the spectral asymmetry of the cone radiation which is observed. There are several factors to be taken into account here: the time variation of the laser pulse in the experiments, the nonlinearity (mentioned above) in the interaction of the waves, and the asymmetry of the propagation of the red and blue components outside the laser beam. It thus becomes necessary to resort to numerical simulation in order to find a quantitative description.

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