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Chemoautocollimation of radiation from a cw CO_2 laser in ethylene

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***Abstract*. The formation of a chemoautocollimation light guide of length approximately 4 cm and diameter 150 μm was observed upon focusing radiation from a 35-W cw gas-discharge CO_2 laser into the ethylene flow. The high-intensity laser radiation in the autocollimation channel initiated ethylene pyrolysis leading to the production of acetylene, and resulted in the visualisation of the channel.**

Keywords: *gas-discharge CO_2 laser, nonlinear effects, autocollimation of a laser beam, chemoautocollimation.*

The self-action of powerful laser radiation caused by the dependence of the refractive index of a medium on the radiation intensity leads to a number of nonlinear effects, in particular, self-focusing and autocollimation of laser radiation. These very interesting effects were mainly observed upon propagation of powerful laser pulses in condensed media [1]. The results of theoretical and experimental studies in this field were analysed in detail in paper [2]. At the same time, the effect of self-action on the propagation of laser beams in gases has not been adequately studied. Worthy of mention are paper [3], in which the autocollimation of a beam from a tunable cw dye laser in sodium vapours was studied, and also paper [4], where self-focusing of radiation from a pulsed CO_2 laser was observed in resonantly absorbing gases BCl_3 and SF_6.

In this paper, we studied the ignition of the ethylene flow in air by laser radiation [5] and unexpectedly found chemo-autocollimation in ethylene of a beam from a cw CO_2 laser of power 35 W only. The scheme of the experiment was as follows. The ethylene flow escaping from a flat fire burner of diameter 6 cm to atmosphere crossed the focal region of the laser beam. A cw CO_2 laser emitted the TEM_{00} fundamental mode. The laser radiation was predominantly determined by the $P(18)$ and $P(20)$ transitions of the 10.6-μm band. The laser radiation was focused by a salt lens with the focal distance 7.5 cm to a spot of diameter approximately 0.3 mm. The focal region in this series of experiments was located at a distance of about 1.0 cm from the burner end, in the region of mixing of the ethylene flow with ambient air. At some power of the laser beam, ethylene was ignited and the front of diffusion flame was formed, which served as a peculiar screen preventing the penetration of atmospheric oxygen into the ethylene flow. Thus, the CO_2 laser beam propagated in fact in pure ethylene, which was confined inside a peculiar chamber, with walls formed by the flame. When the laser beam power was maximal, the 'flash' of autocollimation was observed in the form of a thin luminous filament of length approximately 4 cm and diameter 150 μm (Fig. 1).

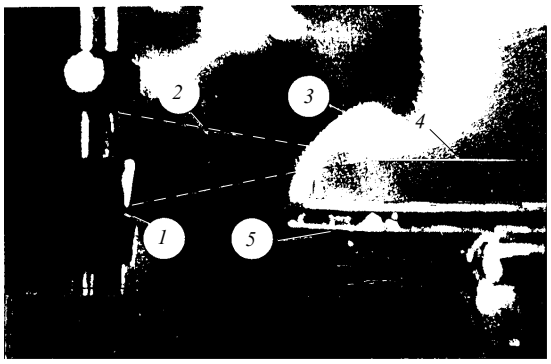

Figure 1. Video picture of the ethylene-flow ignition by a focused beam from a cw CO_2 laser and of the formation of an autocollimation channel in ethylene: (*1*) lens; (*2*) atbitrary profiles of the focused laser beam; (*3*) region of the diffusion flame front; (*4*) autocollimation channel; (*5*) burner.

Autocollimation appeared in the focal region when the invisible beam from the CO_2 laser transformed to a luminous autocollimation yellowish channel. The beam visualisation was indicative of the ethylene pyrolysis within the channel. The ethylene pyrolysis was also confirmed by the formation of carbon particles outside the autocollimation channel over its entire length. Thus, the chemical process allowed the visualisation of the autocollimation channel, which represents in fact a waveguide channel. If autocollimation were absent, the light beam diameter at the channel output would increase from 150 μm to 4 mm.

It is interesting that additional measurements showed that an unfocused laser beam was absorbed in a cell filled with ethylene at atmospheric pressure over a distance of about 2 cm. This raises the question of how autocollimation channels of a substantially greater length are formed. It seems that the propagation of the focused CO_2 laser beam is accompanied by strong heat release, which is determined by the laser radiation intensity and the absorption coefficient of

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ethylene. The latter, according to our estimates, is approximately 1 cm^{-1}. The heating of ethylene in the autocollimation light channel results, first, in tunnelling, i.e., the penetration of the light beam to a greater depth because of a decrease in the density of absorbing molecules. Second, chemical pyrolysis of ethylene molecules proceeding in the autocollimation channel also leads to a partial disappearance of absorbing molecules inside the channel, reducing the attenuation of the laser beam in the channel, which was probably observed in our experiment.

What can we say about the formation mechanism of the autocollimation channel? The refractive index n of nonlinear media is a function of the light intensity. Let us assume that the change in the refractive index n is caused in our case by an increase in the polarisability of vibrationally excited molecules [4], and the dependence of n on the electric field has the form

$$n = n_0 + n_2 E_0^2, \tag{1}$$

where n_0 is the refractive index of the medium in the absence of the field; n_2 is the nonlinear refractive coefficient; and E_0 is the amplitude of the light-wave electric field.

Because the radiation intensity decreases with the distance from the beam axis, the increase in n at the centre of the light channel will be maximal. As the laser intensity increases, the refractive index n in the beam can become so large that the total internal reflection will occur from the boundary, resulting in the formation of a light guide that captures the laser beam. Upon focusing the beam by a lens with the focal distance f, the focusing itself favours the formation of the light guide, and we can readily obtain the relation between the threshold intensity for the formation of the autocollimation channel and the focusing parameters of the laser beam

$$\frac{a^2}{2f^2} = \frac{n_2 E_0^2}{n_0 + n_2 E_0^2} \approx \frac{n_2 E_0^2}{n_0}, \tag{2}$$

where $2a$ is the laser beam aperture; $n_0 = 1.000696$ (hereafter, the refractive indices are presented for the wavelength 589.3 nm). Because n_2 is, unfortunately, unknown, it is difficult to estimate from (2) the threshold power W_t for the laser beam autocollimation for comparing it with the experimental value $W_t = 35$ W. The expression for the threshold power of the laser beam autocollimation obtained from (2) has the form

$$W_t = \pi r_0^2 S = \pi (f\theta)^2 \frac{c E_0^2}{4\pi} = \frac{a^2 \theta^2 c}{8} \frac{n_0}{n_2}, \tag{3}$$

where $r_0 = f\theta$ is the radius of a focal spot; θ is the divergence of the laser beam; S is the laser beam intensity; and c is the speed of light. The value of n_2 can be estimated from expression (3). By substituting in (3) the values $a = 5$ mm and $\theta = 1.5$ mrad and the values of other parameters, we find $n_2 = 8.43 \times 10^{-6}$ cm s^2 g^{-1}. Then, by substituting the obtained value of n_2 into (1), we determine the refractive index n of ethylene inside the autocollimation channel in our experiments. It proved to be equal to 1.002896, which is indeed very high value for gases.

The formation of the luminous light guide in ethylene raises many questions. The most important of them is as follows: if the ethylene pyrolysis occurs due to the ethylene heating in the autocollimation channel, where the laser radiation intensity is $10^4 - 10^5$ W cm^{-2} (which casts no doubt), why the autocollimation channel proves to be so stable? The disappearance of ethylene molecules during pyrolysis should result in the disappearance of the autocollimation channel as well.

It seems that the channel stability is explained by the production of acetylene in pyrolysis, whose refractive index under normal conditions is not only close to that of ethylene ($n_0 = 1.000606$) but also has the close dependence on the laser radiation intensity. In addition, the laser beam frequency and the frequency of the corresponding vibrational–rotational transition in the acetylene molecule can be better matched with increasing n, and the polarisability of excited molecules can be increased, which can strongly affect autocollimation. All these questions require further studies.

Therefore, upon irradiation of ethylene, as an initial nonlinear medium, by a focused beam from a cw CO_2 laser, a sequence of processes occurs. First, autocollimation appears due to resonance absorption of light by ethylene, which results in the formation of a waveguide channel and a drastic increase in the laser beam intensity in the channel. The propagation of intense radiation in the resonantly absorbing medium leads to the ethylene heating in the channel, the intense excitation of vibrational levels, and the ethylene pyrolysis resulting in the production of acetylene, which probably further enhances autocollimation. Therefore, in our opinion, this phenomenon should be called chemoautocollimation, meaning the autocollimation produced due to a chemical reaction induced in the medium.

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