GENERATION OF SUBNANOSECOND PULSES IN NITROGEN LASER-PUMPED TUNABLE DYE LASERS

By

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The temporal behaviour of nitrogen laser-pumped dye lasers was investigated experimentally in order to generate short, subnanosecond, narrow-linewidth pulses. The change of the time position of dye laser pulses was measured by the change of laser wavelength and cavity length. A new type oscillation in dye lasers was studied. This oscillation had cavity round-trip time modulation. By using high-cavity length, tunable subnanosecond pulses were obtained. The intensity competition in two-wavelength lasers can produce subnanosecond pulses with very short fall time.

Introduction

The dynamical investigation of photophysical and photochemical processes demands tunable, narrowband, and very short light pulses. Q switched solid-state lasers, nitrogen laser-pumped dye lasers produce several nanosecond long pulses. Mode-locked dye lasers generate 5—0.2 ps pulses, but they are too expensive. The gap between nanosecond and picosecond pulses could be covered by nitrogen laser-pumped subnanosecond dye lasers.

A great number of methods for producing subnanosecond light pulses have been reported. One of them described in [1—3] is based on relaxation oscillations. In that case, cavities with extremely low decay times are used and, by proper control of pumping level, 0.5—1 ns pulses with 50—500 W peak power were generated. An important disadvantage of this method is that: because of the short cavity no high wavelength selection can be used. 1—1.5 nm spectral linewidths were reported, these are too broad for most applications. Another method for subnanosecond pulse generation using relatively long cavity $L=300$ mm, and high spectral selection has been presented in [4, 5].

Though, to tell the truth, these autors did not distinguish their method from the relaxation oscillations. For a certain spectral range and pumping power, single pulses have been generated, with a pulse duration of 450 ps (fwhm) and 0.002 nm linewidth.

In this paper, we are presenting the results of a systematic investigation of temporal behaviour of nitrogen laser pumped dye lasers. Using these results the conditions for production of single subnanosecond pulses in a long cavity dye laser, and a qualitative explanation of short pulse generation is proposed. A simple and new resonator configuration is presented for very long-cavity dye lasers. As a result of the comprehensive study of the temporal behaviour of two wavelength dye lasers, another simple method is developed for producting short dye-laser pulses.

Experimental arrangement

The most used experimental arrangement can be seen in Fig. 1. Pumping pulses were provided by a nitrogen laser having 200 kW peak power, 7 ns pulse duration (fwhm) operating at 25 pps. A $5 \cdot 10^{-3}$ M/l ethanol solution of Rhodamine 6G and $5 \cdot 10^{-3}$ M/l 7-diethylamino-4-methylcoumarin were used as active material. The dyes were in a 1 cm long flowing dye cell. Five different dye laser configurations were investigated; Fig. 1 shows the most complicated double-wavelength dye laser, and Fig. 2 shows the other four dye laser configurations. In each case, the grating was a 1800 lines/mm ruled grating, and the output coupler was an uncoated flat mirror substrate. The two-wavelength laser had a grating as beam expander [6], and its 0-th order was fed back by another autocollimation grating. DL. 1 is a simple, transversally pumped configuration, DL. 2 uses a grating beam expander; DL. 3 and DL. 4 have an intracavity lens.

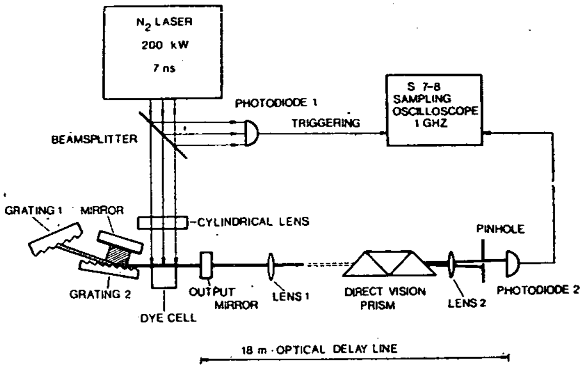

Fig. 1. Experimental arrangement for measuring time position and pulse shape of dye lasers.
Two wavelength dye laser configuration

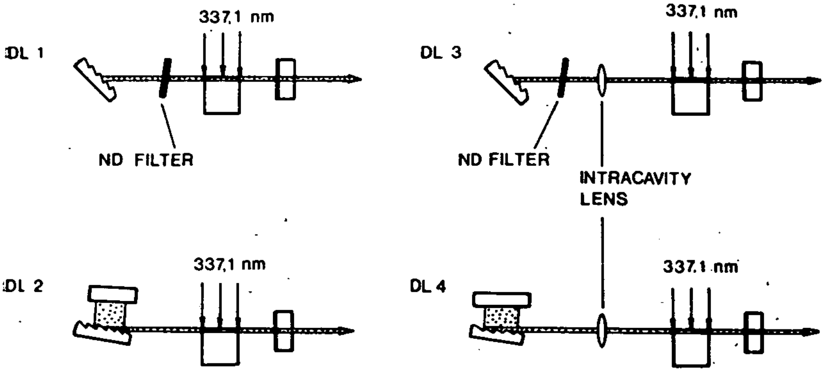

Fig. 2. The used resonator configurations

The intracavity lens was so installed, that the focal plane of the lens coincided with the dye cell side-window; consequently, the grating and the intracavity lens formed a monochromator with the excited region as an entrance and exit "slit". The use of intracavity lens results in higher spectral selectivity and higher efficiency of feedback. With such an intracavity lens DL. 3 and DL. 4 were unstable resonators, but the "walk off" losses were much lower than the other losses of the cavity.

The light emerging from the dye lasers was passed through a lens and a direct vision prism, so that amplified spontaneous emission (ASE), or the outputs of the double-wavelength laser could be selected. At the same time, this spectral selector served as an 18 m long optical delay line. The light was detected by an ITL HSD 1850 biplanar photodiode. A small fraction of nitrogen laser ligth with a TF 50 photodiode triggered the S 7—8 sampling oscilloscope. The rise time of the whole system was 300 ps. The wavelength and linewidth of the dye lasers were measured by a DFS—8 spectrograph and a IT—51 Fabry—Perot interferometer.

Results

Wavelength dependence of the time position of dye-laser pulses

It is well known that the dye-laser pulse apart from some delay, closely follows the shape of the exciting nitrogen laser pulse shape. The delay between the two depends on the pumping power and on the decay time of the cavity. Supposing constant pumping power and constant resonator configuration, there is another important parameter *Viz:* the laser wavelength. The gain curve of the dye depends on the wavelength; with lower gain, the laser is delayed in reaching the threshold and with the maximum gain curve, the laser rises sharply. These dye-laser properties can be predicted by the simple rate equation model [3]. Fig. 3 shows the delay of the dye-laser pulses to be as we calculated, with 100 kW pumping power and $\tau = 10$ ps cavity decay time.

To verify this calculation, the delay have been measured by the use of the DL. 2 resonator configuration. Fig. 4 shows the relative time position of the dye laser

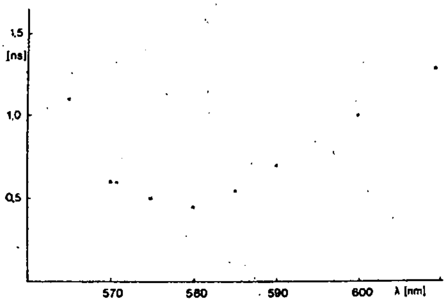

Fig. 3. Calculated time position of dye laser pulses. Parameters: active material Rhodamine 6G, pumping power 100 kW, resonator decay time 10 ps

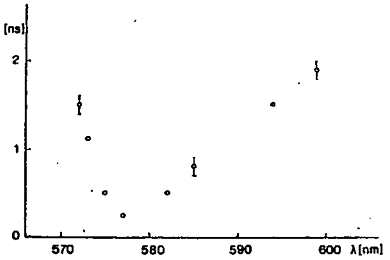

Fig. 4. Measured time position of dye laser pulses. Parameters: as before

pulses. The active material was a Rhodamine 6G solution in ethanol, similar results were obtained for a 7-diethylamino-4-methylcoumarin solution. The qualitative agreement of experimental and theoretical results is satisfactory, because of the simplicity of the model.

The effect of resonator length on the time position of the dye laser pulses

The development of a dye laser pulse begins when the ASE emerging from the dye cell enters the sepctral selector, and a small part of it is fed back to the active region. Using higher spectral selection (*i.e.* beam expander), the distance between the dye cell and the spectral selector could be as much as 10—15 cm, and the above-mentioned round trip takes about 0.8—1 ns. Therefore the time position of the dye laser pulses must depend on cavity length.

Experimental measurements with DL. 2 confirmed this assumption as shown, in Fig. 5. In the course of measurements, the distance between the dye cell and the output coupler was fixed and the cavity length was altered by the movement of the grating. These results show the limits of the simple rate equation model, because when increasing the resonator length the decay time also increases in terms of the model. When substituting increasing decay times for rate equations, the delay decreases contrary to experience.

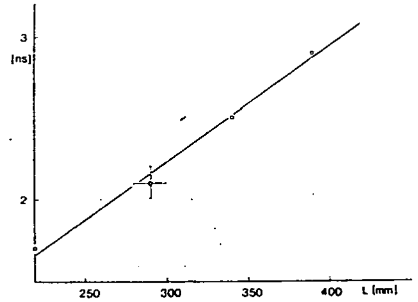

Fig. 5. The relative change of time position of dye laser pulses. Resonator configuration: DL. 2

Oscillations in long cavities

In many cases the shape of the dye laser pulses showed irregular pulsations. These pulsations are not relaxation oscillations, because they could be observed far above the threshold and in a relatively long cavity (30 cm). A typical oscillation is displayed in Fig. 6. The systematic study of such pulses showed that the time separation (t) between subpulses was equal to the cavity round-trip time, and was independent of the pumping power laser wavelength and resonator configuration (see Fig. 7).

The measured points with $L=250$ mm and 400 mm represent different cavity configurations. The measurements described above prove that these oscillations differ essentially from the relaxation oscillations described in [1—3].

To find the origin of this pulsation, the temporal development of the pulses was analised. The temporal development of laser pulses begins with the ASE as previously mentioned. The temporal behaviour of ASE was observed through the output coupler, when the grating from DL. 1 was removed. Fig. 8 shows the ASE pulse as having a short rising half in accordance with the increase of excited-state population. The moment, when the ASE is fed back from the output mirror to the dye cell, the excited state population sharply decreases, and consequently, the level

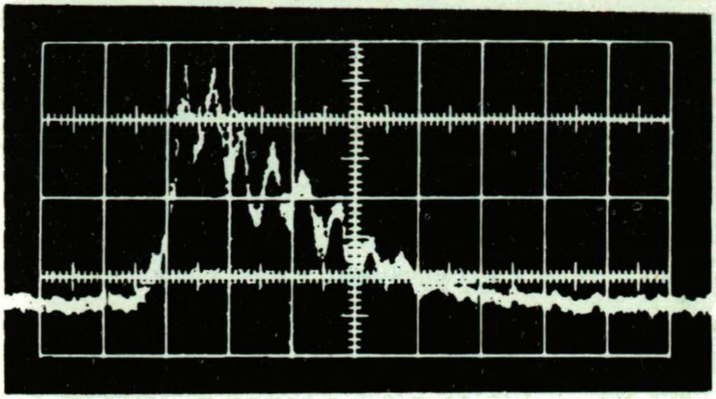

Fig. 6. Oscillations in nitrogen laser pumped dye laser. Horizontal scale: 2 ns/div

of ASE propagating into the direction of the output coupler also decreases. Since the dye pumped strongly, the system would reach the steady state quickly and, therefore, the ASE level would continue to be constant during the remaining part of the pulse. This overshot in the ASE is responsable for the oscillation.

Whether this overshot causes oscillations or not, depends on the efficiency of the feedback of the grating. Here the efficiency of feedback is defined as the photon flux entering the active region from the direction of the grating divided by the photon flux emerging from the active region towards the grating

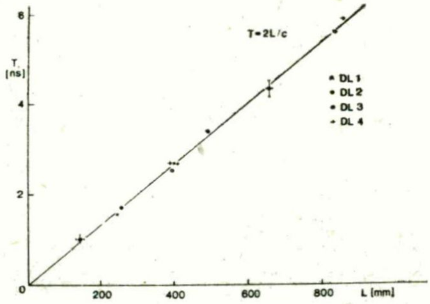

Fig. 7. The time separation between subpulses as a function of resonator length

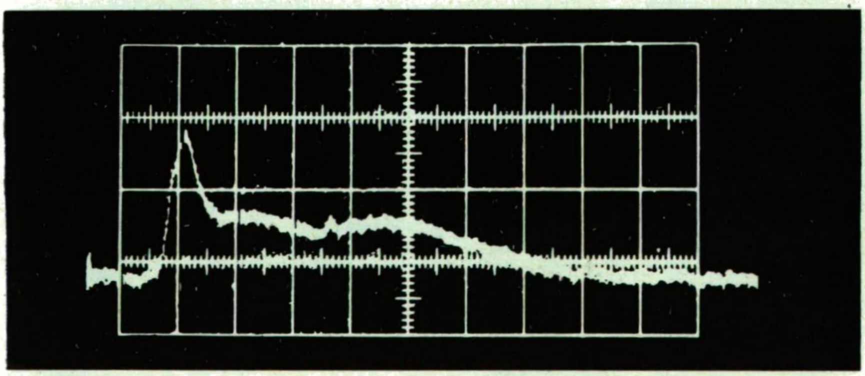

Fig. 8. Temporal behaviour of amplified spontaneous emission in case of one mirror resonator. Horizontal scale 2 ns/div

(*i.e.* spectral selection, transmission of ND filters and the beam expander, reflectivity of the grating and losses due to the divergence of the ASE beam are taken into account). If the intensity of the beam fed back from the grating to the dye cell is so high as to cause deep saturation, no oscillation occurs. (It is well known that the output power of a deeply saturated amplifier depends on the pumping rate alone, and is independent of the input power.)

If the intensity fed back from the grating is so high as to cause partial saturation, the leading part of the pulse will be more strongly amplified than its tail. As a result of subsequent transits through the active region, the initial overshot of ASE gained more, and the tail of the pulse was suppressed. These assumptions were verified experimentally, and as well as theoretically. (See theoretical part.)

Experimentally we changed the feedback in the lasers DL. 1 and DL. 3, inserting a set of ND filters between the grating and the dye cell. With the decrease of feedback, the modulation depth in the dye-laser pulse will grow, and the relative intensity of the pulses follows it by changing (see Fig. 9). Note that the time position of the peaks of subpulses is unchanged. According to our measurements and calculations, the condition of the oscillations is an about 10^{-5}—10^{-6} feedback efficiency. This is why, as a rule in a Hänsch-type laser no oscillations of such kind-were ob-

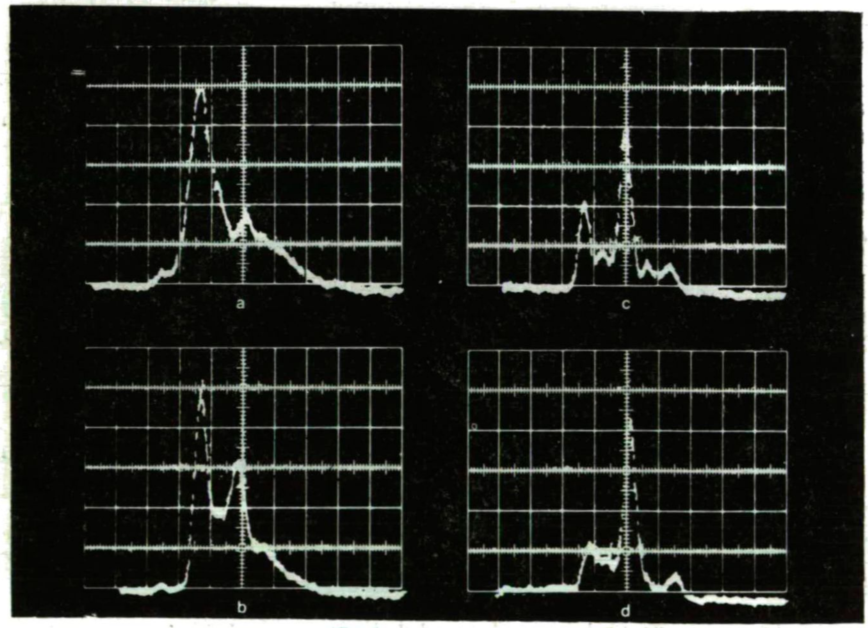

Fig. 9. Pulse shape of dye laser pulses with decreased feedback efficiency

served, since this laser has usually the feedback efficiency of 10^{-3}. A convenient method of the continuous variation of feedback efficiency is the application of a grazing incidence grating. By changing the angle of incidence to the grating, the efficiency of the feedback can be adjusted to such a low value as necessary for the production of the oscillations.

The number of generated subpulses depends on the cavity round-trip time and on the time during which the active medium has a proper gain. With the increase of the cavity length, the number of subpulses may be decreased, and it is possible to generate a single pulse. If the cavity length is increased up to 700 mm for DL. 1 and DL. 2, the laser is about to come up to the threshold. In order to improve the feedback, an intracavity lens was used.

The effect of intracavity lens

The laser arrangement is shown in Fig. 2 DL. 3 and DL. 4. The intracavity lens, as described in the experimental arrangement, formed a monochromator with the active region as an entrance and exit slit. The effectivity of feedback is defined previously. Since both systems, with and without intracavity lens, have the same linear dispersion in the dye cell, it is obvious that the ratio of the effectivities is nearly equal to the ratio of the vertical dimension of the back-reflected of ASE spots *i.e.*:

$$\frac{\eta_{\mathrm{IL}}}{\eta} = \frac{2L\theta}{a}.$$

Where η and η_{IL} are the effectivities of feedback for the simple cavity and cavity with lens, θ is the divergence of the beam emerging from the dye cell and a is the diameter of the active region, L is the distance between the grating and dye cell. Fig. 10 shows the output energies versus cavity length with and without intracavity lens. For longer cavities, the output with intracavity lens is much higher than without lens. It can be seen that the resonator length could be as much as 1 m.

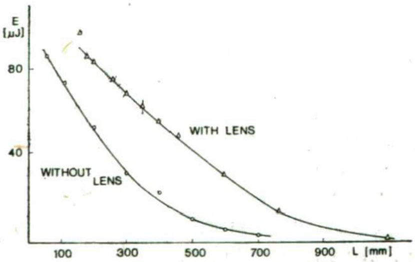

Fig. 10. The effect of intracavity lens on the output energy of dye lasers

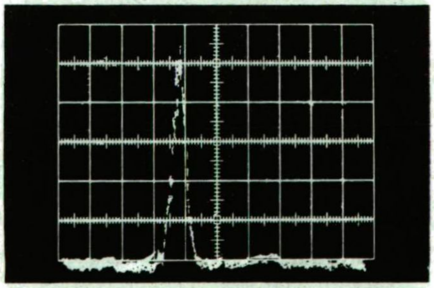

Fig. 11. Subnanosecond output pulse of a 90 cm long cavity dye laser (DL. 4.). Horizontal scale 2 ns/div

By using a 90 cm long cavity with a grazing incidence grating (6 ns round-trip time!) and an intracavity lens, stable single pulses can be obtained. The halfwidth of pulses was 0.55 ns (deconvoluted value), the time bandwidth product was 1.3, and the peak power 8 kW. The shot-to-shot stability was higher than 5 p.c. as seen in Fig. 11.

Temporal behaviour of two-wavelength dye lasers

The two-wavelength operation was achieved by several authors using a double cavity of different type. The relative intensity of the different wavelengths depends on the decay time of resonators and the relative position of wavelegths on the gain curve of the dye. This phenomenon was described by FRIESEM et al [7]. The competition between the independent cavities determines not only the relative intensities but also the temporal characteristics of the two-dye lasers. This temporal behaviour has not been investigated so far.

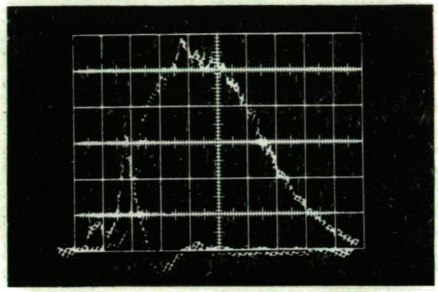

Fig. 12. Time behaviour of a two wavelength dye laser (The outputs were measured separately! 1 ns/div)

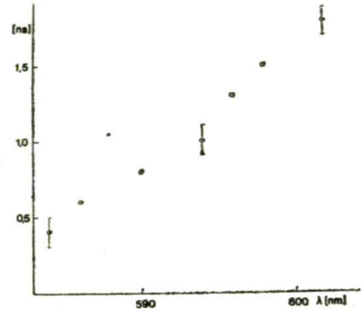

Fig. 13. The halfwidth of the first dye laser pulse as a function of wavelength of second dye laser

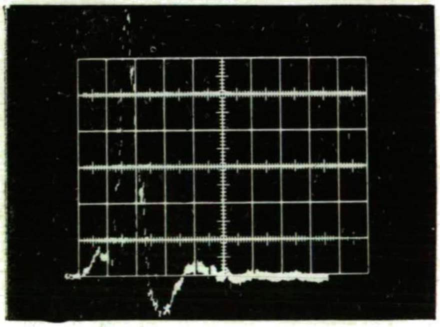

Fig. 14. Subnanosecond pulse of a two wavelength dye laser. Horizontal scale 1 ns/div

A typical pair of dye-laser pulses can be seen in Fig. 12 using the experimental arrangement of Fig. 1. The active material was Rhodamine 6G, and the wavelengths $\lambda_1=574$ nm and $\lambda_2=584$ nm. The most important information of Fig. 11 is that the two lasers do not operate simultaneously. According to the results of the previous section, the shorter cavity operates first. The length of cavities is: grazing incidence grating+mirror: 15 cm, autocollimation grating: 25 cm. A further important characteristic of this system is that the pulsewidth of the first laser pulse depends on the wavelength of the second laser. Fig. 13 shows the pulsewidth of the fixedwavelength first laser for different wavelengths of the second laser. It can be seen that, as the laser wavelength comes near to the maximum gain curve, the second laserises more rapidly, so the halfwidth of the first pulse decreases. Besides the two-wavelength operation, this method allows us to change the pulsewidth continuously and produces subnanosecond operation.

Fig. 14 shows a subnanosecond pulse in case $\lambda_1=574$ nm, $\lambda_2=584$ nm. It is interesting to note that this pulse has not only a sharp rising part but also a sharp fall indicated by the overshot of the oscilloscope.

Conclusions

We have investigated the time behaviour of nitrogen laser-pumped dye lasers. The time position of dye-laser pulses has a strong wavelength dependence. The delay between exciting and dye-laser pulse increases with the increase of cavity length. The origin and most important properties of an oscillation of a new type were investigated. The oscillation has a cavity round-trip modulation time. Using a resonator configuration of a new type and a very long cavity, single 0.55 ns long pulses were generated. It was demonstrated that two-wavelength dye lasers can produce subnanosecond pulses as well.

References

[1] *Lin Ch.:* Journal of Appl. Phys. **46**, 4076 (1975).
[2] *Lin Ch.:* IEEE J. Quant. Electron. **QE—11**, 602 (1975).
[3] *Rácz, B., Zs. Bor, G. Szabó, Cs. Zoltán:* Acta Phys. et Chem. Szeged **23**, 367 (1977).
[4] *Borgström S. A.:* Physica Scripty **14**, 92 (1976).
[5] *Wyatt R.:* Opt. Commun. **26**, 429 (1978).
[6] *Shoshan I., U. P. Oppenheim:* Opt. Commun. **25**, 375 (1978).
[7] *Friesem A. A., U. Ganiel, G. Neumann:* Appl. Phys. Letters **23**, 249 (1973).

ГЕНЕРАЦИЯ СУБНАНОСЕКУНДНЫХ ИМПУЛЬСОВ В ЛАЗЕРАХ, ПЕРЕСТРАИВАЕМЫХ НА КРАСИТЕЛЯХ, ВОЗБУЖДЕННЫХ АЗОТНЫМ ЛАЗЕРОМ

Б. Рац, Ж. Бор, Г. Сабо и Ш. Сатмари

Временная характеристика лазеров на красителях, возбужденных азотным лазером, была исследована для того, чтобы генерировать короткие, субнаносекундные узко-полосные импульсы. Изменение временной задержки импульсов лазера на красителе было измерено изменением длины волны и длины резонатора лазера. Был испытан новый тип осцилляции в лазерах на красителе. Эта осцилляция вероятно модулирована возвратно-поступательным временем резонатора. Получены перестраиваемые, субнаносекундные импульсы, используя длинный резонатор. В лазерах, имеющих две длины волн, конкуренция интенсивностей может привести к образованию субнаносекундных импульсов, именяющих короткие фронты.