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LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

New class of optical apodizing diaphragms on the basis of dyed polymers

S A Volyanyuk, V I Bezrodnyi, E A Tikhonov

Abstract. A new class of apodizing diaphragms, obtained by gradient photobleaching of initially uniform dyed polyurethaneacrylate films by the adapted penumbra technique, is proposed. Two types of intracavity soft apertures are proposed: apodizing selectors that should be used with a separate Q switch, and selectors combined with an optical switch (a single element contains a passive Q switch and an apodizing selector of transverse modes of the cavity). Soft apertures of the first type, produced on the basis of polyurethaneacrylate doped with the dithiobenzyl nickel complex, were used for transverse-mode selection in a Nd³⁺:YAG laser.

Keywords: apodizing diaphragm, dyed polyurethaneacrylate, pulsed Nd^{3+} : YAG laser, super-Gaussian profile.

1. Introduction

The formation of a prescribed spatial profile of laser radiation is required for many problems of nonlinear optics, holography, laser location, and technology. One of the methods of correction for the spatial distribution of laser radiation consists in inserting in the beam the elements whose optical characteristics depend on the radius. Soft or apodizing diaphragms (ADs) belong to such elements.

At present, a number of methods of formation of ADs of different types were proposed and realised [1-15]. They include photographic emulsions with varying blackening [2], dielectric coatings, glass optical filters with deposited metal layers of varying thickness [3, 15], dye solutions in cells with varying thickness [3], Pockels and Faraday cells with nonuniform electric and magnetic fields [5], elements based on absorption induced in transparent materials by ionising radiation [6], and optical elements based on attenuated total reflection [7].

Studies in this area are of considerable importance because many soft apertures do not satisfy all requirements imposed on such elements. ADs based on photographic emulsions with varying blackening, which are the first elements that were successfully used for laser beam apodization, have the lowest damage threshold and a poor optical

Received 25 October 2000; revision received 21 February 2001 *Kvantovaya Elektronika* **31** (5) 456–460 (2001) Translated by A N Kirkin quality, which is caused by a nonuniform thickness of photographic plates, the presence of grains in a photographic emulsion, scratches, etc [1]. ADs based on single-layer dielectric coatings have a low contrast. Such ADs are used in practice for correcting a light beam after its passing through an additional soft aperture having a high contrast.

For this reason, new techniques of production of ADs are developed and previously proposed techniques are improved [8-12]. In Ref. [13], the holographic technique for manufacturing mirrors with a varying reflectivity is developed. This method makes possible simultaneous control of frequency and spectral width of laser radiation. The authors of [14] propose a new apodizing element, which represents a photopolymer holographic phase plate for its use in a cavity of a diode-pumped Nd: YAG laser.

Depending on the problem, ADs can be placed at the output of a master oscillator or in different parts of an amplifying system [1], cutting out from a beam being amplified the part with a nearly uniform distribution, or inside a cavity for selecting the fundamental transverse mode [8–10, 13, 14].

The use of ADs is one of the simplest methods for eliminating spatial nonuniformities caused by Fresnel diffraction from apertures of a laser system [3]. The Fresnel diffraction with intensity gradients is responsible for the selffocusing of a beam as a whole and small-scale focusing [16], which cause damage of laser optics.

The use of soft apertures in laser systems imposes on them the following requirements: (1) the presence of a nearly rectangular spatial transmission profile, which changes, however, at the edges so that diffraction intensity spikes are suppressed (for instance, such properties are provided by the super-Gaussian profile [1]); (2) the presence of a specified (as a rule, higher than 100 for intracavity applications) transmission contrast; (3) the optical damage threshold (ODT) of an AD should exceed the intensity of laser radiation transmitted through it; and (4) the technology should provide the desired service life of an AD.

In this paper, we solve several interrelated problems: we develop the technology of fabrication of ADs on the basis of dyed polyurethaneacrylate, study spatial profiles of ADs manufactured by this technique, and study specific features of selection of transverse modes by soft aper-tures in comparison with mode selection provided by hard metal diaphragms.

2. Method of AD formation

We fabricated ADs for a Nd³⁺:YAG laser by using a triplex design in which a polyurethaneacrylate (PUA) film

S A Volyanyuk, V I Bezrodnyi, E A Tikhonov Institute of Physics, National Academy of Sciences of Ukraine, prosp. Nauki 46, 03039 Kiev, Ukraine

 $100-500 \ \mu\text{m}$ thick doped with the dithiobenzyl nickel complex (DBN) was formed during photopolymerisation between glass substrates of good optical quality. This material and the technology of triplex production were proposed earlier for passive Q switches of solid-state lasers [17]. We extend the range of application of this material and propose to use it for the fabrication of ADs.

The absorption spectrum of DBN is presented in Fig. 1b. The absorption spectrum of the dye overlaps the emission frequencies of lasers working on the ${}^{4}F_{3/2} - {}^{4}T_{11/2}$ transition (1.06-µm region) of neodymium in all known matrices. DBN has a high resistance to photochemical bleaching in the fundamental absorption band at 1.06 µm and is rather easily decomposed under irradiation in the near UV region, where electronic transitions to higher states are located. The photochemical properties of DBA in PUA were found to be favourable for the fabrication of ADs by the gradient photobleaching technique. The optical characteristics of PUA obtained by the radical photopolymerisation of oligourethaneacrylate are presented in Ref. [17]. PUA has a transparency window in the range from 0.4 to 1.5 um (Fig. 1a), which allows one to use this material for the fabrication of ADs in this spectral range by using different dyes. The elasticity of PUA provides a high ODT of this polymer matrix [18, 19].



Figure 1. The transmission spectrum of PUA (a) and the absorption spectrum of DBN (b).

Dye diffusion in PUA is one of the processes which can change the initial gradient profile of an AD. We have shown for coumarin 7 [20] that the diffusion of dye molecules in elastic PUA is rather slow. For a concentration of 10^{-3} mol litre⁻¹ at the centre of the diffusion region (a film 170 µm thick), the diffusion coefficient at 300 K is (8.3 ± 0.4) $\times 10^{-11}$ cm² s⁻¹. The initial concentration of coumarin 7 in the bulk of the dyed polymer (at a considerable distance from the diffusion boundary) was 2×10^{-3} mol litre⁻¹. The diffusion coefficient increased with increasing film thickness (for coumarin 7, by an order of magnitude). The dye diffusion in polymers was found to have a dispersion nature, i.e., decreased with time [21]. The diffusion coefficient also depends on the size of a diffusing molecule and, therefore, it is different for various dyes. For DBN (with molecular weight of 629.55), diffusion into PUA is slower than for coumarin 7 (with molecular weight of 333). This provides the diaphragm lifetime required for practice.

The AD lifetime can be estimated by the formula $t_{ad} = l^2/D$, where *l* is the diffusion length and *D* is the diffusion coefficient for dye molecules. For l = 1 mm and $D = 10^{-11}$ cm² s⁻¹, the time of efficient exploitation of ADs (taking into account only the diffusion of dye molecules) is 33 years. Thus, optical, photochemical, and diffusion properties of this composition are favourable for the use in the given technology.

The calculation of the diffraction pattern for a soft aperture shows that the apodizing ability of an AD (i.e., its ability to suppress diffraction intensity spikes simultaneously with realisation of a large filling factor) increases on passing from the Gaussian to the super-Gaussian profile. The super-Gaussian function, which is used for the description of AD profiles, has the form

$$T(r) = T_0 \exp\left[-\left(\frac{r}{R}\right)^N\right].$$
(1)

Here, T(r) is the transmission as a function of the distance r from the aperture centre; T_0 is the transmission at the aperture centre; R is the distance from the aperture centre to the point where $T = T_0/e$; and N is the order of the super-Gaussian law.

To form an AD with the super-Gaussian transmission profile by dye photobleaching in PUA films, we adapted the penumbra technique [22], which was earlier used for the fabrication of ADs based on silver-halide photographic films. The initial technique used an incandescent lamp with a circular hard aperture and an opaque screen in the form of a disk as a light source. We modified this method by using a UV lamp with an amplitude mask, which was placed at a certain distance from a polymer film. The amplitude mask represented a circular hard aperture made in a metal foil. The desired spatial transmission profile of an AD was obtained by photobleaching a dyed plane-parallel polymer layer by the UV emission.

To average the irradiation dose in the azimuthal direction, the initial dyed triplex with an amplitude metal mask was placed in a special device rotating about its axis. As noted above, the DBN photostability upon excitation to high electronic states is low. The dye photodecomposition is accelerated with time because of reactions involving decomposition products containing excited DBN molecules.

The irradiation parameters (the exposure time, the position of the initial composition relative to the light source, and the use of amplitude masks of different diameter) were chosen empirically. For a triplex placed at a distance of 7 cm from the UV lamp, a noticeable dye decomposition (an increase in transmission by 10%) was observed after 7-h exposure. By using moderate exposure times (about 30 h for triplexes with an initial transmission of

To form transmission profiles approximated by super-Gaussian functions, we increased the exposure time, all other conditions being the same. The technique chosen by us imposes no substantial restrictions on the profile being formed and the operating wavelength of an AD and provides fabrication of ADs with the prescribed contrast.

By using this technique, we fabricated ADs of two types: diaphragms with a super-Gaussian absorption gradient at the edges and total transmission at the centre and diaphragms combined with a passive laser Q switch. In the second case, the transmission at the aperture axis has a maximum value, but does not reach 100 %, and decreases towards the aperture edges according to a super-Gaussian law. A nonlinear element of this type can be simultaneously used for mode selection and Q switching. It is expected that lasing in a system with such an element will begin at the axis on the fundamental transverse mode and spread in the radial direction during the development of a 'giant' pulse, filling the active medium and preventing the appearance of higher-order modes. Earlier, similar elements based on LiF crystals with F_2^- -colour centres were proposed in Ref. [23] (see also [8]).

An additional advantage of the use of an initially dyed polymer film placed between two optical surfaces for manufacturing ADs is that the film surface in this system is protected from mechanical damage (in an AD based on a photographic film with varying blackening, scratches and dust particles adhered to the emulsion deteriorate its optical quality). Moreover, a relative simplicity of the fabrication and a small size of ADs proposed here favour their extensive use in laser systems.

3. Measurement and analysis of AD transmission profiles

The schematic of the setup for measuring the transmission profile of ADs is shown in Fig. 2. The 1.06- μ m radiation from a 20-Hz diode-pumped Nd³⁺: YAG laser *1* passed through a focusing system 2, an optical fibre 3 and a scattering system 4, was partially absorbed by an AD 5, and detected by a CCD camera 6. The output signal was digitised by a frame grabber and a computer 7.



Figure 2. Schematic of the setup for measuring profiles of apodizing diaphragms.

The profiles obtained by our technique were nearly super-Gaussian and were approximated by functions (1). The order N of the diaphragm transmission profile ranged from 2 to 20. By increasing the diameter of a hard aperture mask (with the exposure time and the distance between the mask and the UV lamp being fixed), we can obtain various

profiles from the Gaussian (with N = 2) to the super-Gaussian one with N = 20. By increasing the exposure time, with the amplitude-mask diameter being unchanged, ADs with the desired contrast ratio I_{max}/I_{min} can be obtained. Note that the contrast as low as 1.43 was found to be sufficient for efficient operation of intracavity ADs.

Fig. 3 presents examples of normalised profiles for some soft apertures manufactured by us. Let us compare the super-Gaussian profile with N = 6, R = 2.6 (curve 3) and the Gaussian profile with N = 2, R = 2.6 (curve 1). The super-Gaussian transmission distribution is more uniform than the Gaussian one. As shown in Ref. [1], for the super-Gaussian intensity distribution, the filling factor at the output of a soft aperture is higher than for the Gaussian distribution. Therefore, the output energy of a system in the first case is higher. This means that an AD with N = 6 provides a more efficient use of the section of a laser rod.



Figure 3. Normalised profiles of ADs with the exponential parameters N = 2 (1), 4 (2), 6 (3), 12 (4), 18 (5), and 20 (6) for soft apertures with profile radii of 2.6 (1, 3, 5), 3.6 (2), 2 (4), and 1.5 mm (6).

We studied ADs with a smooth gradient of absorption coefficient, which were combined with a passive laser Q switch and ADs that were not combined with a switch. The dependences of their transmission on the coordinate are presented in Fig. 4.



Figure 4. Profiles of ADs combined with laser switches for N = 18 (1) and 4 (2) and of the AD that is not combined with the optical switch for N = 6 (3).

4. Results of the use of ADs in laser cavities

Soft apertures with a smooth gradient of absorption coefficient were used in a cavity of a Nd³⁺:YAG laser simultaneously with a passive Q switch. It is known that for the diffraction pattern formed by a soft aperture with radius a be smoothed at a distance of z' from the aperture (in linear media) the changes in the Fresnel number $F = a^2/\lambda z'$ caused by the irregularity of the edges or their spreading should be of the order of unity [1], i.e.,

$$\Delta F = 2a \frac{\Delta a}{\lambda z'} = \frac{2\Delta a}{a} F \approx 1.$$
⁽²⁾

It follows from this formula that the required spreading of the edges of a soft aperture is $\Delta a \approx a/2F = \lambda z'/2a$.

However, it is likely that this principle is invalid when an AD is used inside a cavity for selecting the fundamental transverse mode. We studied ADs in a plane-parallel Fabry–Perot cavity (a cavity 110 cm long with a Nd³⁺ : YAG crystal 6 mm in diameter) using an LBA-300PC (SPIRI-CON) analyser of laser beams. Our measurements showed that the ADs provided efficient selection of higher transverse modes for *F* in the range from 1 to 14 and ΔF in the range from 0.4 to 8.9. We also studied ADs in a cavity with a passive *Q* switch, whose initial transmission was 11 %. Under such conditions, lasing on the fundamental mode was obtained by using the AD with a = 3.5 mm, N = 6, F = 10, $\Delta a = 1.5$ mm, $\Delta F = 4$, and contrast ratio of 1.43.

Fig. 5 presents the laser radiation profiles measured in the near-field zone (immediately behind the output mirror) in the laser without an AD, with an intracavity AD, and with a hard diaphragm 2.7 mm in diameter. Figs 5a-5chave the same scale, the frame size is $6.336 \text{ mm} \times 5.940 \text{ mm}$. Our studies showed that the soft aperture with a super-Gaussian profile suppressed diffraction intensity spikes and provided selection of the fundamental transverse mode of Nd³⁺:YAG laser emission. In the case of a hard intracavity diaphragm (for instance, a hole in a metal foil) with the same Fresnel number, the Airy diffraction pattern was observed in the laser intensity distribution. This field profile no longer corresponds to the fundamental transverse mode and is caused by the Fraunhofer diffraction from a circular aperture. Multimode laser radiation with a complex structure is also obtained when the mode selection is performed using a hard aperture of a smaller diameter compared to that of the AD. The analysis of Fig. 5 shows that Fresnel numbers required for selecting the fundamental transverse mode using intracavity ADs differ from Fresnel numbers for hard diaphragms.



Figure 5. Near-field distributions at the output of a Nd^{3+} :YAG laser without an AD (a), with an AD in the cavity (b), and with a hard aperture 2.7 mm in diameter (c).

In Res. [8, 23], the application of ADs combined with passive laser switches provided a decrease in the angular divergence of single-pulse lasers and an increase in their brightness due to the mode selection in the regime of nonlinear transmission. A detailed study of mode selection by soft diaphragms will be carried out elsewhere.

5. Conclusions

The study of transmission profiles obtained by using ADs fabricated by photobleaching of dyed polyurethaneacrylate films by the penumbra technique showed that this technique imposes no substantial restrictions on the profile being formed and enables one to obtain ADs with the desired contrast ratio. Soft apertures based on dyed PUA films provide selection of the fundamental transverse mode of laser radiation at large Fresnel numbers.

The use of hard diaphragms, even of smaller diameter (compared to that of apodizing diaphragms) leads to the appearance of higher transverse modes in laser radiation. In the case of a soft diaphragm, the efficiency of formation of the fundamental transverse mode (the ratio of energy obtained in the fundamental transverse mode to the maximum possible energy of the fundamental transverse mode for a hard aperture) at large Fresnel numbers is 4.3.

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