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# Broadband optical limiting in thin nanostructured silicon carbide films and its nature

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ABSTRACT

Broadband optical limiting of the ns laser radiation in visible and near IR spectrum range in nanostructured films of silicon carbide $\beta$-SiC(3C) studies are presented. The nature of the optical limiting mechanisms is discussed. The contributions of the optical nonlinear absorption and nonlinear scattering are determined and measured. It is shown that the nonlinear absorption is related to the two-step carrier excitation into the conducting band through the local levels in the band gap followed the absorption by nonequilibrium carriers. The mechanism of the nonlinear scattering is Rayleigh at its origin and arises as a result of difference of the refractive indices caused by the giant nonlinear refraction in nanocrystals of SiC and environment. It is shown that the Z-scan-like technique with the variable aperture size allows for combining contribution of both the nonlinear absorption and the nonlinear scattering mechanisms to increase the efficiency of the optical limiting.



## 1. Introduction

Among optical limiting devices for power laser radiation special interest is devoted to those which could be used in the optical communication area as an optical switching, laser beam control as well as sensitive detectors and eye protection when working at extremely aggressive conditions i.e. high and low temperatures, high level of light and radiation power, and chemical atmosphere [1,2]. Materials with corresponding linear and nonlinear optical properties should be available for such limiters. One of such nonlinear materials is the indirect band gap semiconductor silicon carbide (SiC) which is characterized by high optical and mechanical strength, thermal stability, chemical inactivity and devises on the basis of SiC can be used in the outer space [3–5]. Some nanostructured SiC polytypes, in particular, α-SiC (6H), may be characterized by second order nonlinear susceptibility $\chi^{(2)}$ [6,7]. Different SiC polytypes have wide band gap which changes from 2.3 eV for β-SiC(3C) to 3.3 eV for α-SiC (2H) and allow for controlling high power laser beams in wide spectral range. It was shown in our previous papers [8,9] that SiC, in particular, α-SiC (21R), (27R) and β-SiC (3C) thin nanostructured films are characterized by high third-order optical nonlinear susceptibility $\chi^{(3)}\sim 10^{-5}$ esu. In the samples of these polytypes we observed optical limiting effect measured at laser wavelengths $\lambda=532$ nm and 1064 nm [10]. Now the nature of the optical limiting and conditions for the maximum efficiency in the nanostructured SiC thin films is not clarified. Thereupon it should be noted that the effect of optical limiting observed in some direct band gap semiconductors is related to two-photon absorption [11,12].

In our paper [10] the optical limiting effect was observed using Z-scan-like technique with an open aperture. However, in such a configuration the technique reflects only nonlinear absorption and does not allow for observing possible small angle nonlinear scattering and estimating its contribution into the optical limiting effect. Yet this contribution should be high enough taking into account large nonlinear refraction in the nanostructured SiC.

In this paper we present the results of study of the optical limiting in the $\beta$-SiC(3C) nanostructured thin films obtained by means of the Z-scan-like technique with a variable aperture and a nanosecond laser pulses at $\lambda_1=1064$ nm and $\lambda_2=532$ nm. For the theoretical analysis of experimental results a mathematical model for two configurations of the experimental scheme with open and variable aperture was developed. Using the model we analyzed our experimental results and obtained value of the nonlinear absorption and nonlinear scattering contribution into the optical

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limiting effect.

## 2. Experiment

Nanostructured silicon carbide films were produced by the original technique of direct deposition of silicon and carbon ions on quartz or sapphire substrates at a relatively low (less than 1500 °C) temperature [13]. This technology allows for controlling the process of nanocrystalline film formation, and thus to obtain samples with different polytypes of silicon carbide $\alpha$-SiC (21R, 27R), $\beta$-SiC (3C), and percentage of crystalline phase (from 20% to 100%) [14].

Silicon carbide single crystals [15], has a sharp fundamental absorption edge in the range of photon energies 2.4–3.4 eV, depending on the polytype. The absorption spectra of the nanostructured silicon carbide, we have synthesized, have extended 'tails' of the states in the visible and near-IR spectrum ranges (Fig. 1), indicating the presence of nanoparticles with different local levels in the band gap of the material. These are primarily surface defects of a developed general nanoparticle system surface, which itself is an origin of crystal structure defects, generating the so-called Tamm levels. In addition, these states in the band gap can be associated with the presence (along with the main polytype of silicon carbide nanomaterials) of some percentage of other polytypes as well as byproducts of synthesis in nanostructured samples. Thus, according to photoelectron spectroscopy [16], the nanostructured films, apart from heterobound SiC atoms, also contain homobound atoms of silicon Si–Si and carbon C–C. In this case, at the substrate temperatures of 700–950 °C, silicon, not bound to carbon, crystallizes into a nanocrystalline phase with particles similar in size to silicon carbide. Also the band energy parameters dependence on nanocrystalline sizes is important to nanostructured SiC absorption end blurring [17]. It should be noted that the sufficient absorption in the visible and near-IR regions, we observed in the spectra of silicon carbide nanoparticles, indicates that the concentration of these states is very high and they can form an impurity band inside the band gap. Heat treatment of the synthesized nanostructured silicon carbide films by annealing at 1000 °C followed by etching leads to a significant depletion of these states in the band gap [10].

Optical limiting in the nanostructured samples of SiC was studied using the experimental scheme shown in Fig. 2, which is similar to that used in [18] but with some modification suitable to our purposes. It should be noted that the sample is placed in the focus of the lens L1 and does not move along beam propagation

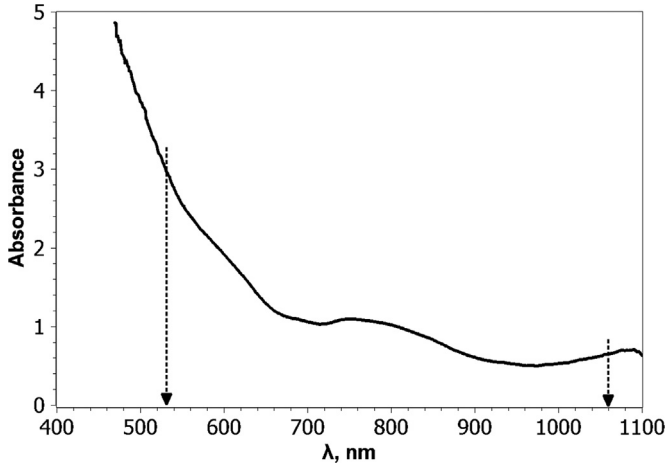


**Fig. 1.** Absorbance spectrum of the $\beta$-SiC(3C) polytype sample. Laser wave lengths are shown by the arrows.

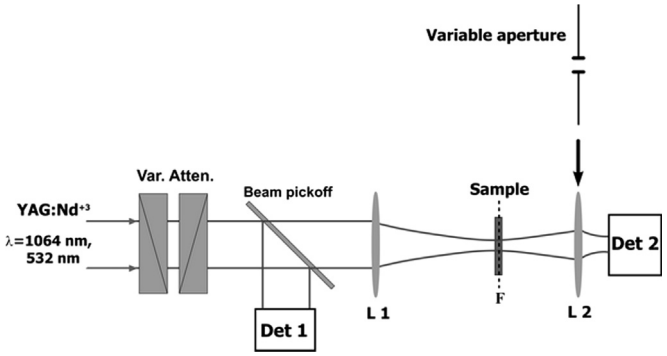


**Fig. 2.** Experimental set-up.

direction ($Z$ axis). The depth of the focus or the confocal parameter for Gauss laser beams is $b=\frac{2\pi w_0^2}{\lambda}$, where $w_0$ is the radius of the beam waist in the focus where the laser wave front is plane. In our case the parameter $b$ is about 3 mm for the $\lambda=532$ mm. That is why to place a sample with the thickness of 0.45 μm in the area with the dimension of 3 mm is not too complicated. In addition it is strongly controlled. We place the second lens L2 in the scheme to collect the light passing through the sample (case 1 "open aperture"). The case 2 of the experimental studies is related to the variable aperture, when the aperture is placed instead of the lens 2.

The technique with the open aperture allows for measuring the nonlinear absorption more correctly in comparison with the case of variable aperture. However, it is not sensitive to small angle nonlinear scattering. At the same time, the configuration with the variable aperture allows for separating the relative contribution of the nonlinear absorption and the nonlinear scattering into the extinction coefficient and measures their values.

In the experiment we used a single-mode $Q$-switched: YAG:$Nd^{+3}$ laser with a pulse duration $\tau_1=20$ ns at the fundamental wavelength ($\lambda_1=1064$ nm) and $\tau_2=15$ ns at the second harmonic wavelength ($\lambda_2=532$ nm).The laser beam is focused on a sample by a lens L1 with a focal length $F=12$ cm. Radiation transmitted through the sample is collected by a similar lens L2 and registered by a recording system in the technique with the open aperture. In the measurement under consideration this lens was replaced by the variable aperture before the recording system. The input laser radiation intensity was controlled by an attenuator consisting of a $\lambda/2$ plate and a Glan prism. The laser pulse energy at the input and output of the circuit was measured using an automated multi-channel recording system. The results are averaged over 20 laser pulses at a pulse repetition rate of 1 Hz.

## 3. Mathematical model

To analyze the experimental data theoretically with respect to the sample transmission, we should first construct a mathematical model. The model is usually validated by comparing the results of computational and physical experiments. It is known that in an optical medium with the linear absorption, the relative change of light intensity $\Delta I/I$ in the samples along the direction of propagation $z$ is proportional to the change of distance $\Delta z$:

$$\frac{\Delta I}{I} = -\alpha_0 \Delta z, \tag{1}$$

where $\alpha_0$ is the linear absorption coefficient.

In semiconductors, the linear absorption $\alpha_0$, along with the processes of two-photon (or two-step) absorption $\beta$ and nonlinear scattering $\gamma_S$ are present. Therefore, in Eq. (1), instead of the