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Hybrid metal polymer as a potential active medium of an optoacoustic generator

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A hybrid material was studied, consisting of polydimethylsiloxane and silver nanoparticles distributed throughout its volume, its optical and thermodynamic characteristics were calculated for different volume fractions of silver content. It is theoretically shown that this material with a volume fraction of silver of about 30% can be used as an active medium for an optoacoustic transducer with an operating frequency range of about 10 MHz.

Keywords: ultrasonic generator, polydimethylsiloxane, hybrid material

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The operation of an optoacoustic generator is based on expansion and contraction of its active region due to heating as a result of absorption of laser pulses and subsequent relaxation [1,2]. Systematic studies of optoacoustic methods have been performed in the USSR since the 1970s by a research group led by L.M. Lyamshev and K.A. Naugol?nykh at the Andreyev Acoustics Institute and other research groups [3,4]. Materials with suitable optical, thermodynamic, and mechanical characteristics need to be used in an instrument to maximize the efficiency of conversion of the laser radiation energy to the mechanical energy. First, complete absorption of the laser radiation at the lasing wavelength should be implemented; second, the material should have a high coefficient of thermal expansion; third, the active region should cool sufficiently fast for the system to relax before the absorption of the next pulse [5].

Since the active medium material needs to satisfy several requirements simultaneously, it is very hard to find a single versatile medium. However, it is possible to engineer a hybrid material that combines the advantages of its components and negates their disadvantages. Hybrid materials are now widely used in photonics as light-emitting materials [6,7], in solar panels [8], and to control terahertz radiation [8–10].

Polydimethylsiloxane is often used as an active medium material for optoacoustic generators, since it features an exceptionally high coefficient of thermal expansion [11,12]. However, it does not absorb laser radiation efficiently on its own. Therefore, one needs to combine it with a metal layer [13] or metallic nanoparticles [14]. Optoacoustic generators of this design have already been constructed and reported in literature [15]. The restriction on the operating wavelength (lasers of the blue-green range need to be used to ensure fine absorption in the metal) is the primary technical disadvantage of such designs. It was demonstrated in [5] that the use of structures with a Tamm plasmon provides an opportunity to achieve complete absorption at the wavelength of GaAs semiconductor lasers, which are the lasers of choice due to their availability, high power, and great modulation capabilities.

Polydimethylsiloxane with integrated silver nanoparticles was chosen as the hybrid material with its parameters to be modeled in the context of its possible use as the active medium of an optoacoustic generator. Since polydimethylsiloxane is a heat-insulating material, the redistribution of temperature in a solid layer proceeds at a very low rate. The characteristic time of temperature equalization in a material layer may be estimated using the following formula [5]:

$$\tau = \frac{d^2}{2D}, \tag{1}$$

where d is the layer thickness and D is the temperature conductivity of polydimethylsiloxane. Thus, the characteristic time of temperature distribution throughout a layer with a thickness of 500 nm is on the order of 1000 ns. This makes proper operation at frequencies above 1 MHz infeasible, while the generation of ultrasonic waves in the $2-29$ MHz range is needed in medicine and non-destructive testing. A hybrid material providing uniform heating of the organic layer throughout its entire volume was chosen to address this problem.

In order to design a structure with a Tamm plasmon based on a hybrid material, one needs to know its effective optical characteristics (such as the refraction and absorption coefficients). The Maxwell Garnett formula, which relates the permittivity of a mixture (ε_{mix}) to the volume fraction of inclusions (f) and the permittivities of components, may be used to characterize a two-component heterogeneous

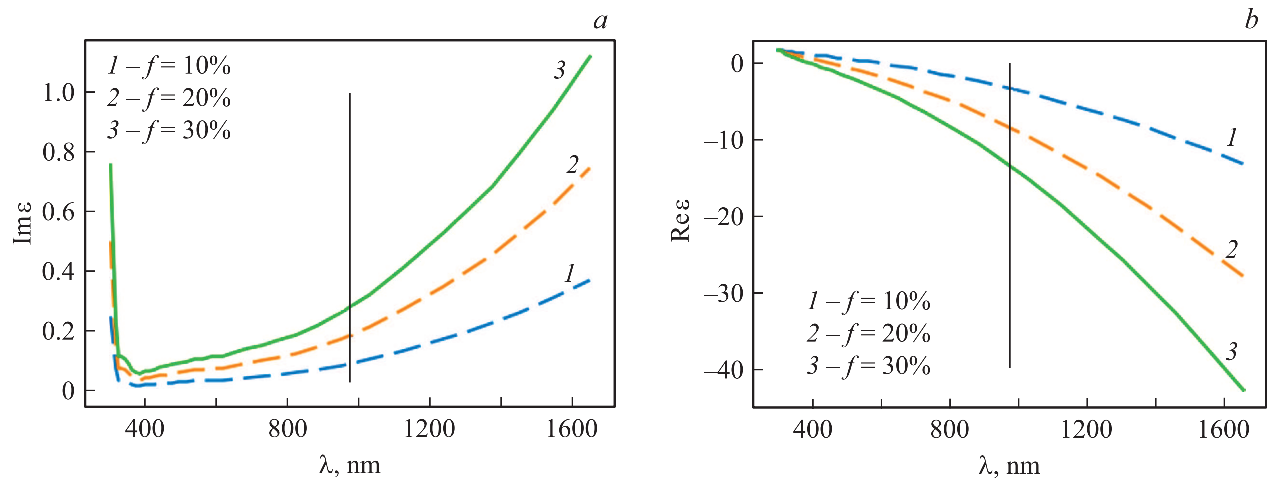

Figure 1. Spectra of imaginary (*a*) and real (*b*) parts of the effective permittivity of the hybrid material with different volume fractions of silver nanoparticles. The vertical line marks the operating frequency of semiconductor lasers based on GaAs (980 nm).

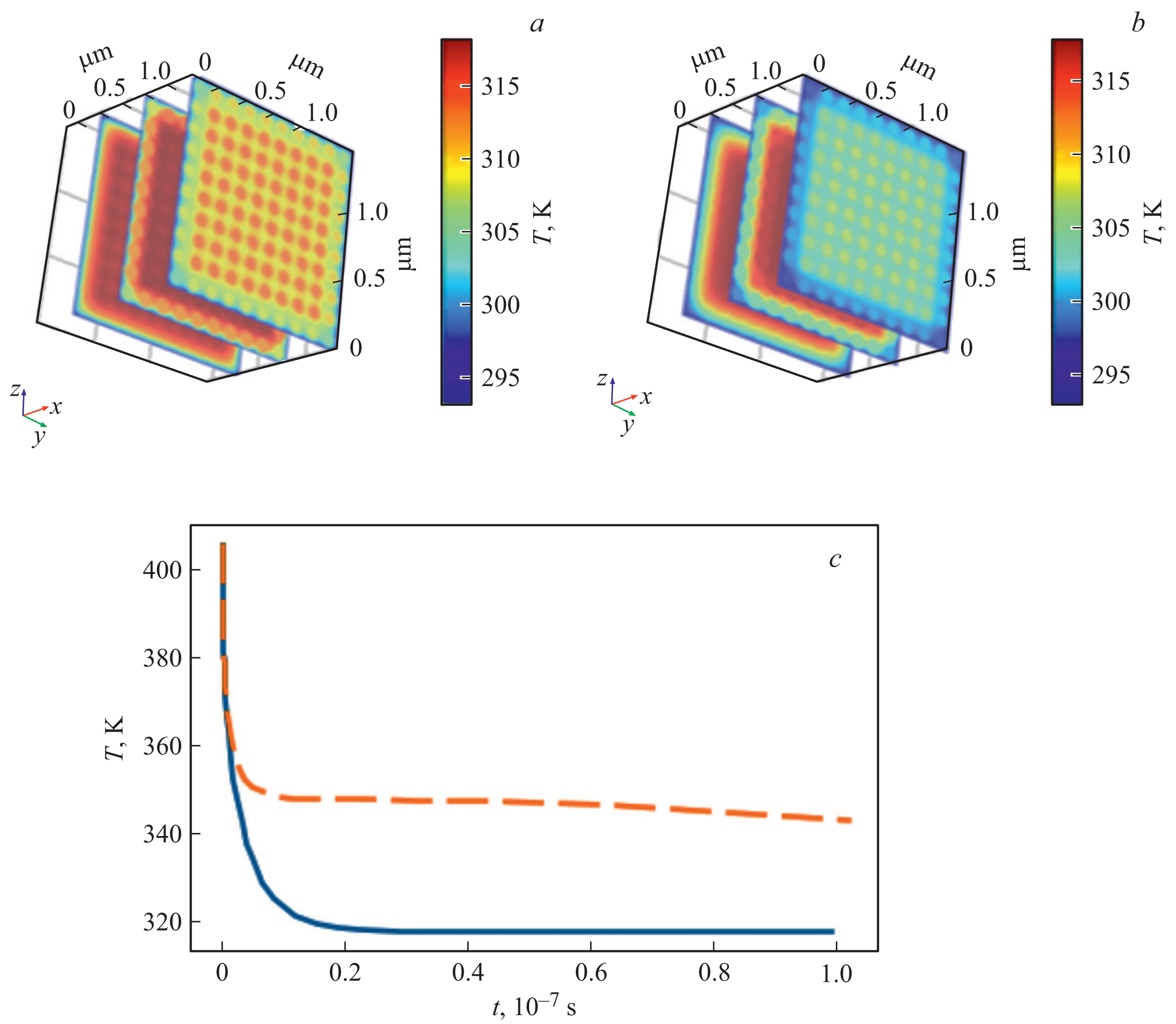

Figure 2. Temperature distribution in the structure with a volume fraction of nanoparticles of 29% within 40 (*a*) and 60 ns (*b*). *c* — temperature reduction with time in the process of cooling of materials with a volume fraction of particles of 17% (solid curve) and 29% (dashed curve).

Dependences of thermal conductivity A, temperature conductivity D, thermal capacity c, density ρ, and real n and imaginary k parts of the refraction coefficient on percentage f of silver nanoparticles

f, %	A, W/(m · K)	D, 10^{-7} m^2/s	c, J/(kg · K)	ρ, kg/m^3	n ($\lambda = 980$ nm)	k ($\lambda = 980$ nm)
10	0.26	1.7	790	1917.5	0.024	21
12.5	0.285	1.8	715	2155.63	0.027	25.3
15	0.332	2.18	655	2393.75	0.028	26.1
19	0.4	2.48	580.4	2774.75	0.031	29.8
29	0.55	3.1	365	3727.25	0.038	37.2
100 (Ag)	430	961	235	10490	0.04	6.9624
0 (PDMS)	0.16	1.1	1460	965	1.3951	6.9624

system with the volume fraction of one component being no higher than 1/3:

$$\varepsilon_{mix} = \varepsilon_{\text{PDMS}} \left[1 - \frac{3f(\varepsilon_{\text{PDMS}} - \varepsilon_{\text{Ag}})}{2\varepsilon_{\text{PDMS}} + \varepsilon_{\text{Ag}} + (\varepsilon_{\text{PDMS}} - \varepsilon_{\text{Ag}})} \right], \quad (2)$$

where $\varepsilon_{\text{PDMS}}$ is the permittivity of polydimethylsiloxane and ε_{Ag} is the permittivity of silver. This formula was used to calculate the spectra of real and imaginary parts of the effective permittivity that are presented in Fig. 1. The data for silver and polydimethylsiloxane were taken from experimental studies [16].

The active medium of an optoacoustic generator with wide application possibilities should provide for efficient energy conversion in different modes of ultrasonic examination with the use of both wide-interval pulses and a continuous modulated wave (e.g., CW Doppler ultrasonography that is being introduced as a method for monitoring the parameters of blood flow of unstable patients [17,18]). Proper temperature relaxation of a system is no less important than its efficient heating. Simulations were performed in Comsol Multiphysics to estimate the time of cooling of the hybrid metalorganic material.

Cooling of the structure in air was simulated for nanoparticles with a radius of 50 nm integrated into a polydimethylsiloxane layer with volume fractions of 17 and 29%. The number of nanoparticles in both calculations was 1000. Cooling and the redistribution of temperature in the structure were simulated on the assumption that metallic nanoparticles were initially heated to 405 K (maximum heating that is not critical for polydimethylsiloxane) and that the initial temperature of polydimethylsiloxane and the environment is room temperature (293.15 K). It is reasonable to set the operating temperature range of an instrument closer to the boundary established by the organic component of the hybrid material, since a greater contrast with room temperature provide for more efficient cooling. Fig. 2, a, b present the temperature distribution in the structure with a volume fraction of nanoparticles of 29% within 40 and 60 ns. Fig. 2, c shows the time dependences of the maximum temperature within the structure. It can be seen that the structure with a volume fraction of nanoparticles of 17% relaxes to a much greater extent (to 320 K) in 100 ns than the structure with a volume fraction of 29% (350 K).

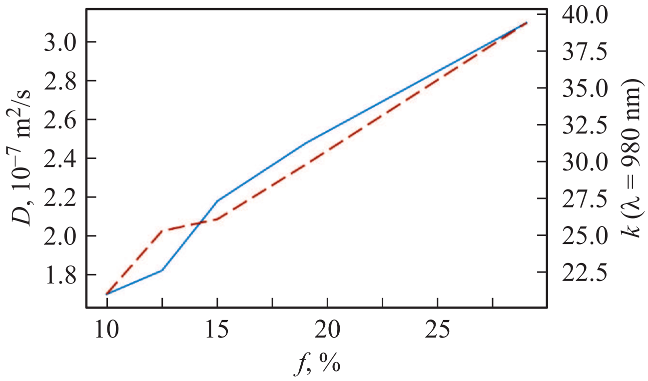

Figure 3. Dependences of effective temperature conductivity D (solid curve) and imaginary part k of the refraction coefficient at a wavelength of 980 nm (dashed curve) on the volume fraction of silver nanoparticles.

The propagation of a heat flux through a hybrid material layer was simulated in Comsol Multiphysics, and the values of thermal conductivity for different volume fractions of silver nanoparticles were determined this way. The values of the key effective optical and thermodynamic parameters of the hybrid material are listed in the table. Fig. 3 presents the dependences of the temperature conductivity and the imaginary part of the refraction coefficient on the percentage of silver. Since both quantities increase with the volume fraction of silver nanoparticles, one may conclude that a material with a fraction of nanoparticles close to 30% (percolation threshold) is better suited for use as the active medium of an optoacoustic transducer. However, one should also bear in mind that the system should have sufficient time to relax at the intended operating frequencies.

Thus, it was demonstrated that a hybrid material consisting of a polydimethylsiloxane matrix and integrated metallic nanoparticles may be regarded as an active medium for an optoacoustic generator, since is absorbs at the wavelength of a semiconductor laser and provides sufficient temperature relaxation for the instrument to operate at frequencies on the order of 10 MHz. The effective optical and thermodynamic properties of the material, which may be used to design the instrument, were calculated.

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Conflict of interest

The authors declare that they have no conflict of interest.

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