Generation of Directed X-ray Radiation in Layered Structures of Opal Matrixes and Piezoelectric Materials for Application in Medicine

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Abstract

Experimental modeling of processes and devices for generating directional X-ray radiation induced by pulsed laser action on samples of layered structures with opal matrix (SiO_2) - piezoelectric ($LiNbO_3$ or $BaTiO_3$) in the form of plates and films was carried out: The composition and structure of opal matrixes and nanocomposites based on them, as well as the experimental conditions (measurements were carried out in the temperature range from 300 K to 100 K) influenced on the characteristics of X-ray beam. The possibility for generation of the pulsed X-ray radiation with an angular divergence of ~ $1 \cdot 10^{-3}$ rad was demonstrated. The studied materials are promising for solving a large number of practical problems, for example, in medicine during endoscopic investigations. Placing opal matrixes at the output of an optical fiber with laser radiation makes it possible to deliver X-ray radiation directly to the irradiated object. By connecting a fiber optic system to a Raman spectrometer, the effectiveness of X-ray exposure on a given object could be observed and controlled.

1. Introduction

Due to the presence of forbidden photonic bands and regions of anomalous increase in the density of photonic states, photonic crystals are of interest from the point of view of controlling the characteristics of laser radiation and obtaining new effective sources of electromagnetic waves. Photonic crystals based on OM artificial material, which is a regular packing of spherical amorphous SiO_2 nanoparticles of equal diameter, are promising for practical applications [1, 2]. Of great interest is the application of opal matrixes for the generation of X-ray radiation (with photon energies from 10 eV to several MeV) in the development of devices for medicine [3, 4]. Solving the problem of increasing the technological characteristics of X-ray sources by creating fundamentally new designs will make it possible to develop subminiature X-ray sources for lithotripsy and endovascular surgery. The goal of this research was to study directed X-ray radiation induced by pulsed laser excitation of materials based on opal matrixes.

2. Experimental Technique

Opal matrix samples were synthesized by hydrolysis of a mixture (suspension) of orthosilicic acid tetraester ($Si(OC_2H_5)_4$) with ethanol (C_2H_5OH) in the presence of ammonium hydroxide (NH_4OH) according to a previously developed method [5-7]. Formed samples with a volume of up to 10 cm^3 were subjected to drying and heat treatment in a drying cabinet at 420 K for 24 hours and in a muffle furnace at 970 K for 10 hours, after which they were strengthened at a temperature of ~1400 K and a pressure of 0.2 MPa. To obtain bulk material with specified diameters of spherical SiO^2 particles in the range of 200-400 nm, the concentration of $Si(OC_2H_5)_4$ was changed. A series of OM samples were manufactured with a diameter (d) of spherical SiO2 particles from 200 to 350 nm (Δd for each series < 4%) (Fig. 1). The strengthened OM samples had contact pads in the shape of a circle with a diameter of up to 0.3 d in the area of contact of spherical particles.

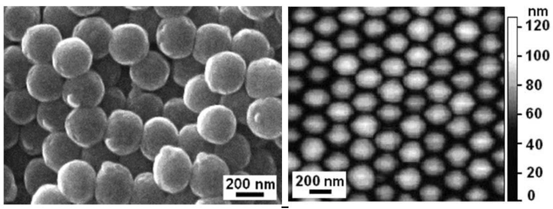

Figure 1. Structure of opal matrixes: scanning electron microscopy of the cleavage surface of a bulk sample and atomic force microscopy of the growth surface.

The packing of spherical SiO_2 particles forms a lattice with tetrahedral and octahedral cavities, which conventionally consist of spheres inscribed in the cavities and spaces connecting them. An ordered system of communicating cavities occupies ~26% of the OM volume. The maximum size of free space at the junction of tetrahedral and octahedral cavities is ~0.39d.

3. Impact of Laser Radiation on Opal Structures

Opal matrix (OM) samples consisting of spherical SiO_2 nanoparticles with d ≈ 250 nm, when exposed to one pulse of laser radiation, demonstrated a radiation strength of 550 GW/cm^2, while for opal matrices with d ≈ 300 nm the radiation strength was 350 GW/cm^2. The radiation strength of OM samples and nanocomposites based on them, depending on their composition and structure, changed in the range of 100-600 GW/cm^2, which is significantly greater than that of crystalline or fused quartz. In photonic-phonon materials, which include OM, the energy of powerful optical action by laser pulses is converted into acoustic vibrations with amplitudes that are non-destructive to the material. The energy spectra of the X-ray radiation produced when the OM is exposed to pulsed laser excitation were measured. Based on the spectral data, it was concluded that the induced X-ray radiation is a low-intensity soft X-ray radiation with a photon energy of 0.08-2.47 keV and a wavelength of 15.2-0.5 nm [3, 4]. The fabrication conditions and structure of the OM (thickness, high-temperature annealing, filling of intersphere cavities), as well as the experimental conditions (obtaining X-ray radiation in transmission or at an angle of 90°, cooling the OM) influence the measured characteristics.

In Figure 2 the schema of a device for generating directional pulsed X-ray radiation under laser irradiation is presented [4]. The device includes a ruby laser operating in Q-switching mode (1). Laser radiation (2) using lenses (3) was focused on a composite structure (4, 5) consisting of an OM plate (5) bonded to plates of piezoelectric materials (4). The laser beam was directed parallel to the contact surfaces of the plates. The composition structure is placed on a massive copper plate (8), which provides heat dissipation. In some experiments, a copper plate (8) with a fixed composite structure was placed in a cuvette with liquid nitrogen (9). The X-ray film was fixed at a distance of 5-20 cm from the opal matrix. One of the options for placing an X-ray film or gamma spectrometer (7) for recording X-ray radiation (6) is given at the scheme.

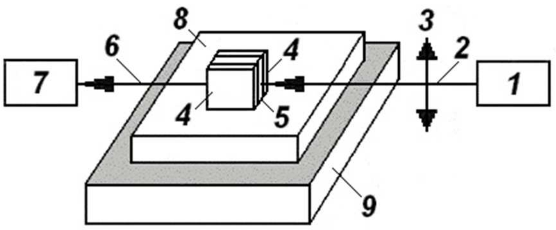

Figure 2. Schema of a device for generating and recording directional pulsed X-ray radiation: 1 – source of laser radiation; 2 – laser beam; 3 – optical system for focusing laser radiation; 4 – plates made of piezoelectric material; 5 – OM plate; 6 – X-ray radiation; 7 – X-ray recording system (cassette with X-ray film or gamma spectrometer); 8 – copper plate; 9 – heat removal container with liquid nitrogen.

The place where the film was exposed to a single X-ray pulse was a small-diameter bright point with a characteristic spatial distribution. Changing the intensity of laser radiation did not lead to a change in spot size. Removing the cassette with X-ray film from the investigated sample at a distance of 50 to 200 mm did not lead to a significant change in the size and relative intensity of the recorded signal, which indicated a small solid angle of radiation for a single pulse. Without the plates based on piezoelectric materials, X-ray radiation was generated, the direction of propagation of which was not reproduced and a large beam divergence was recorded. The application of a compositional structure significantly reduced the scatter of single X-ray pulses, so that the generated X-ray radiation was directed. As a result of the action of pulsed laser radiation (nanosecond duration range) on the OM, which represents a three-dimensional photonic-phonon medium, acoustic radiation in the range of 5-15 GHz and pulsed X-ray radiation were generated. At laser radiation intensities exceeding the threshold for the onset of acoustic oscillations and the accompanying glow of the sample, X-ray radiation was recorded on the X-ray film in the form of a separate illumination. The composition of OM and nanocomposites based on them influenced the result of interaction of laser radiation with them. The generation of X-ray radiation with a small angular divergence ($\sim 1 \cdot 10^{-3}$ rad) took place when OM samples whose cavities were filled with different substances were exposed to pulsed laser radiation with a power in the range of 0.25-10 GW/cm^2. The angular divergence of X-ray radiation of OM:Ni nanocomposites at a temperature of 100 K and a laser radiation power of 9.9 GW/cm^2 was $\sim 1 \cdot 10^{-3}$ rad. The size of Ni crystallites in the OM:Ni nanocomposite was 20–43 nm, with ~20% cavities filling. By filling the structural nanocavities of OM with fluids with different refractive indices and using matrices with different sizes of spherical particles, we can effectively control the functional characteristics of OM. The X-ray divergence depended on the piezoelectric material and the measurement temperature and when using $LiNbO_3$ plates ($LiNbO_3$/OM/$LiNbO_3$ structure) was $<1 \cdot 10^{-3}$ rad. The best results were obtained with mechanical attachment of opal matrix plates and $LiNbO_3$. When using $BaTiO_3$ plates, regardless of the method of their attachment to the OM plate (silicone grease, glycerin, mechanical clamp) and the sample temperature (100 - 300 K), the X-ray divergence was $\sim 1 \cdot 10^{-1}$ rad. When using piezoquartz plates (quartz-SiO_2/OM/quartz-SiO_2 structure) with a small electromechanical coupling coefficient ($k^2 \approx 0.05$ - 0.1%), the X-ray divergence under the same conditions was ~1.5 rad.

The high efficiency of converting high-power laser pulses of one type of energy (optical radiation) into long-term dissipation with formation of the other types of oscillations in non-crystalline dispersed structures creates the opportunity for the application of optical media based on opal matrixes.

4. Conclusion

Nonlinear optical effects arising in opal matrixes under pulsed laser excitation have been studied. Opal matrixes were used to generate acoustic waves and X-ray radiation under laser irradiation. The results of measuring the characteristics of X-ray radiation induced by pulsed laser action on samples based on opal matrixes in the form of plates and films are presented. The conditions for using opal matrixes and nanocomposites based on them as an effective source of X-ray radiation under pulsed laser excitation have been determined. The use of composite structures with opal matrixes and piezoelectric materials made it possible to create directed X-ray radiation.

Reducing the spread of generated X-ray pulses was achieved by manufacturing a sample of an opal matrix in the form of a plate in contact with plates of piezoelectric materials cutted from single crystals of piezoelectrics with $k^2 > 0.2\%$, which made it possible to create a preferred direction for acoustic waves having photon-phonon interaction with X-ray radiation.

The X-ray divergence depended on the piezoelectric material and the measurement temperature and when using plates of $LiNbO_3$ and $BaTiO_3$ ($LiNbO_3$/OM/$LiNbO_3$ and $BaTiO_3$/OM/$BaTiO_3$ structures) was ~ 10^{-3} rad.

The studied materials are promising for solving a large number of practical problems, for example, in medicine during endoscopic investigations. Placing opal matrixes at the output of an optical fiber with laser radiation makes it possible to deliver X-ray radiation directly to the irradiated object. By connecting a fiber optic system to a Raman spectrometer, the effectiveness of X-ray exposure on a given object could be observed and controlled.

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