ESRF	**Experiment title:** STRUCTURAL CHARACTERIZATION OF GAIN MEDIA FOR RANDOM LASERS AND FLUORESCENCE EMITTERS ON THE BASE OF NANOPOROUS SILICA GLASSES	**Experiment number**: 26-02/582
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Scientific background. The experiments aimed on the structural characterization of a new type of photonic glasses based on the porous silicon oxide spheres. The diameters of spheres were varied from 250 to 1200 nm with monodispersity better than 5%. The disordered media of SiO_2 spheres both amplify and scatter light [1] and are used for random lasers, fluorescence emitters and others photonics and nanoelectronics applications. The mechanism of the light amplification in the random lasers has found to be a multiple coherent light reflection on the inhomogeneous structures, which results in formation of accidental but circularly locked trajectories. The real interest to the random lasers appeared recently in connection with the demonstration of the light amplification effect on the random inhomogeneous photonic structures [2-6]. These structures are built of disordered in space but *homogeneous* spherical particles of one or sometimes two different diameters of the nanoscale sizes (photonic glasses). We propose a new unique solution to create working media for the random lasers – to use disordered photonic media (photonic glass), made of spherical particles of silicon dioxide SiO_2, having a regular internal substructure of nanochannels with a diameter of 3-6 nm. That allows one to significantly increase the resonant interaction of the electromagnetic field with an active medium of the photonic glass by filling the nanochannels inside the spherical particles SiO_2 with organic fluorescent colored liquids. This will result in the improvement of the characteristics of the random lasers on the qualitative level.

Samples and experimental techniques. In the experiments we used two techniques μXRD and SAXS to study two length scales of the sample of order of 0.5-1.2 microns and of 2-6 nm, respectively. The ordinary SAXS measurements with a short sample-detector distance of 1.5 m were performed prior to the μXRD measurements. Later on the μXRD setup at the BM-26B (DUBBLE) beamline was constructed, which is unique and not (yet) available anywhere else. A set of beryllium compound refractive lenses was used to focus the beam at the detector which stays 8m far away of the sample. We used the Pilatus detector with the pixel size 175x175 microns square for the SAXS measurements and the 9x9 microns square Photonics Science detector in the μXRD setup.

Photonic glasses were synthesized on the basis of the monodisperse spheres SiO_2 with the internal regular structure of channels with diameter of 3-6 nm. Mesoporous silica spheres are prepared on the basis of procedure described in [7]. The method involves the polycondensation of a silica source (tetraethyl orthosilicate (TEOS), 98%, Aldrich) in the presence of a surfactant (cetyltrimethylammonium bromide, CTAB, 99.9%, Aldrich) in an aqueous solution. As a result, submicron monodisperse spherical particles were obtained. Depending on the content and synthesis details, the samples show optical properties either of a photonic crystal or a photonic glass. This transition is of interest itself but it also allows to determine the value of spatial inhomogeneity, which is crucial for the highly effective light amplification. The films with the thickness from 5 to 50 μm and square area of 5x10 mm^2 were used for the x-ray studies.

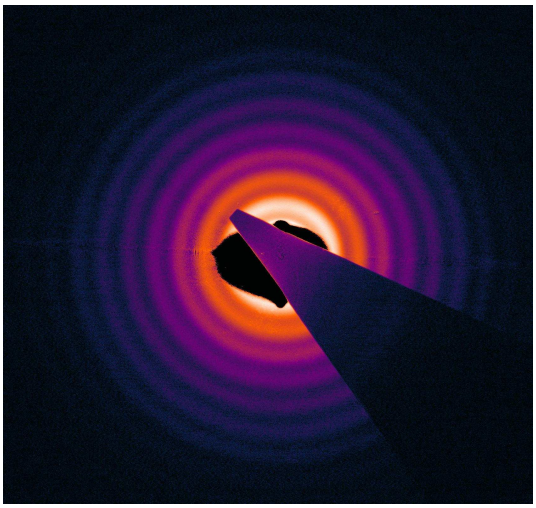

Figure 1. Microradian diffraction pattern taken for the GMT25az photonic glass.

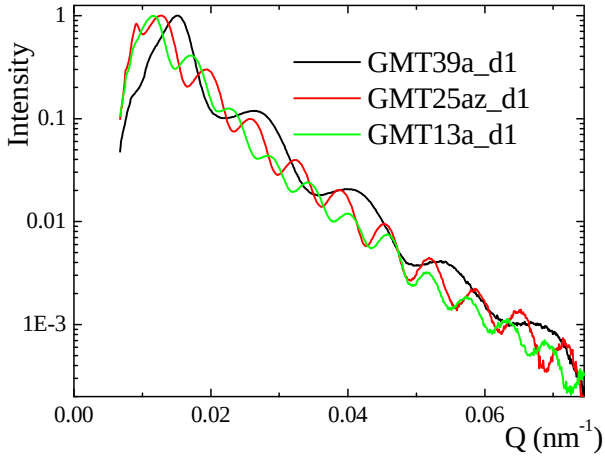

Figure 2. Q-dependence of the scattered intensity for photonic glasses on the base of the regularly speckled spheres with different diameters.

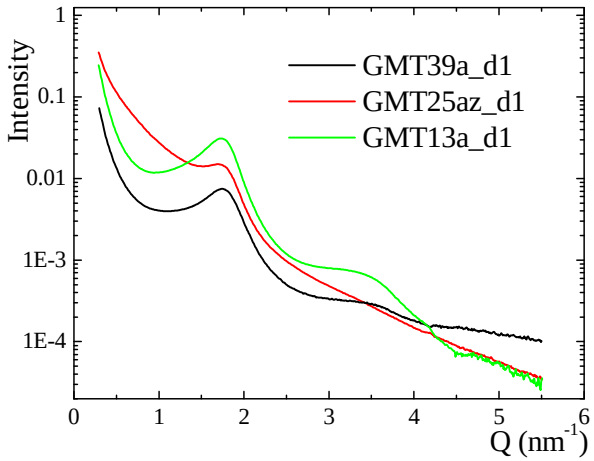

Figure 3. Q-dependence of the scattered intensity on the regular internal substructure of nanochannels in SiO_2 spheres with different diameters which form the photonic glasses.

Results. The microradian diffraction pattern shown in Figure 1 was taken from photonic glass GMT25az grown from mesoporous spheres of amorphous silica, using sedimentation on a substrate of fused quartz 0.7 mm thick. The film thickness was 6 layers. Similar patterns were obtained for all investigated samples regardless on the content and technology method of synthesis. The difference was only recorded in the different periodicity of the rings disposed on a equal distance from each other. This is a fingerprint of the disordered system giving a formfactor of the sphere in the scattering pattern. Q-dependences of the scattered intensity for samples with different spheres diameters are presented in Fig. 2. Accounting for the complete disorder of the samples, we calculated diameters of the spheres, which the photonic glasses are made of. The results correspond to D = 450 ± 10 nm, 960 ± 10 nm and 1080 ± 10 nm for samples of GMT39a, GMT25az and GMT13a series, respectively.

The periodicity of the regular internal sphere substructure of nanochannels can be determined from the positions of the Bragg reflections (Fig. 3) and equal to 3.55 ± 0.05 nm for all investigated samples. Note the asymmetric shape of the diffraction peaks, which may be the result of either noticeable asymmetric dispersion of nanochannels periodicity, or due to exotic internal nanochannel structure deviating from the hexagonal 2D order in the arrangement of nanochannels to the radial 3D one.

Thus, from the experiments we obtained detailed information 1) on the structure and the long-range periodic order of the inner superstructure of SiO2 spheres (ordering of nanopores with diameter of 3-4 nm), 2) on the disorder and short-range periodic order in the photonic glasses itself.

References

1. V. S. Letokhov, Sov.Phys., JETP **26,** 835 (1968).
2. P. D. Garcia, Advanced Materials, **19**(8), 2597 (2007).
3. S. Wiersma, Nature Physics, **4**, 359 (2008).
4. J. S. Fallert, Nature Photonics, **3**, 279 (2009).
5. H.E.Tureci, Science, **320**, 643 (2008).
6. P.D. Garcia, Advanced Materials, **22**(1), 12 (2010).
7. Hernando, J.Phys.Condes.Mater., **48**, 9455 (1999).