

## Modeling of X-ray Diffraction by Carbon Nanotubes and Interpretation of Diffractometry of the Films Deposited in Tokamak T-10

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**1. Introduction.** The x-ray diffractometry of the films deposited in the vacuum vessel of tokamak T-10, carried out at the Kurchatov Institute Synchrotron Radiation Source (wavelengths  $\lambda=0.1$  nm, 0.0464 nm), as well as using a conventional x-ray tube source ( $\lambda=0.154$  nm), has shown [1,2] the presence of a wide peak at low scattering angles  $\theta$  (namely, at  $q \sim 1$  Å<sup>-1</sup> in Fig. 1, where  $q = |\vec{k}_s - \vec{k}_i| = \frac{4\pi}{\lambda} \sin(\theta)$  is modulus of scattering wave vector) which roughly correspond to fluctuations of elementary scatterers' density in the range  $\sim 10$  nm. This peak appeared to be not explainable by contributions of typical impurity (including graphite) polycrystals and most popular nanostructures like fullerenes or carbon nanotubes (CNTs). The breadth of the peak indicates on its statistical origin.

Here we report on numerical modeling of angular distribution of x-ray diffraction by the carbon nanostructures and present a (non-unique) solutions of an inverse problem of reconstructing the distribution of carbon nanostructures over a fixed set of structural patterns to interpret experimental data [1,2]. The set includes (i) CNTs of various diameter, length, wall number, and chirality, (ii) fullerenes C<sub>60</sub> and bigger spheres and ellipsoids, (iii) graphite nanocrystals and graphene sheets, (iv) toroidal graphitic carbon.

### 2. Comparison of observations with diffraction by various carbon nanostructures.

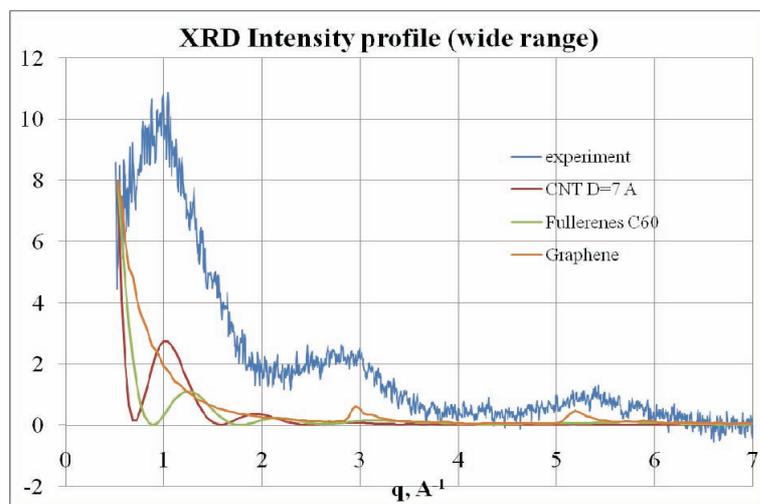


Fig. 1. Comparison of observed angular distribution  $S_{\text{exp}}(q)$  with possible contributions of CNTs (diameter  $D=7$  nm, averaged over chiral angle), C<sub>60</sub>, graphene sheet. The orientation of nanostructures is homogeneously random. The computed curves are normalized to be everywhere not higher than  $S_{\text{exp}}(q)$ . For nanotubes,  $S_{\text{theor}}(q)$  (computed per unit length) with increasing length  $L$  is close to limiting case  $L=\infty$  for aspect ratio  $L/D$  of few units. In this paper, we give results for  $L/D=5$ .

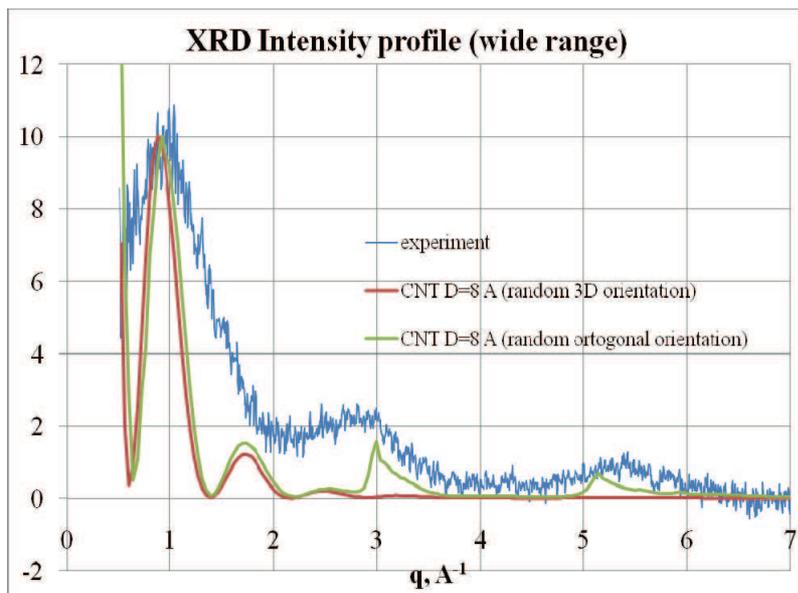


Fig. 2. Comparison of observed angular distribution  $S_{\text{exp}}(q)$  with possible contributions of CNTs of diameter  $D=8 \text{ nm}$ . The orientation of nanostructures is homogeneously chaotic either in 3D or 2D case (CNTs are orthogonal to the incident wave). The computed curves are normalized to be not higher than  $S_{\text{exp}}(q)$  in the vicinity of the peak at  $q \approx 1 \text{\AA}^{-1}$ .

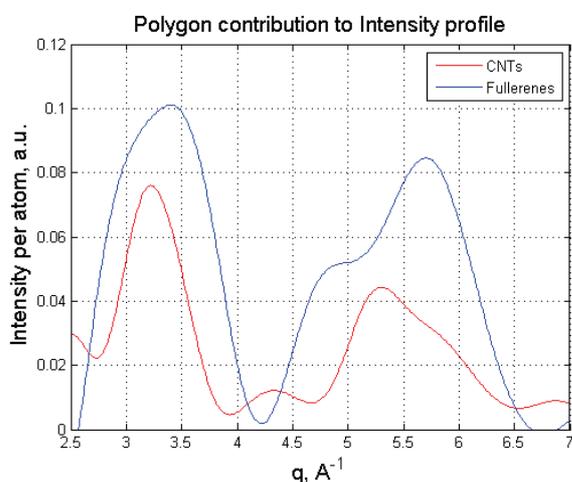


Fig. 3. The approximate universality of contribution of polygons (hexagons and pentagons) in a curved graphene of random orientation, calculated as a difference of scattering by - randomly-oriented CNTs and continuous-wall tubes of similar diameter,  $D=7 \text{ nm}$ , (red), and - randomly-oriented fullerenes  $C_{60}$  and continuous-wall sphere of  $D \approx 7 \text{ nm}$  (blue).

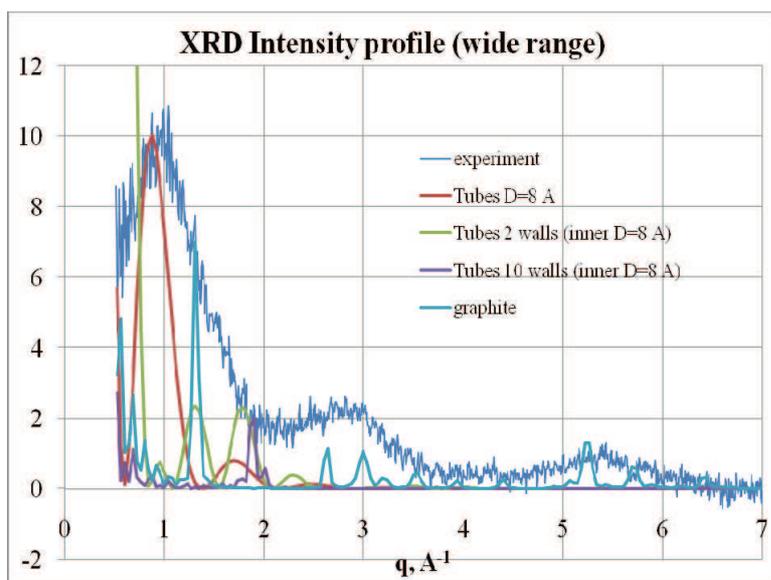


Fig. 4. Comparison of observed angular distribution  $S_{\text{exp}}(q)$  with possible contributions of randomly-oriented single-wall and multi-wall continuous tubes, and graphite nanocrystal (20 sheets). The computed curves are normalized to be not higher than  $S_{\text{exp}}(q)$  in the close vicinity of the peak at  $q \approx 1 \text{\AA}^{-1}$  and to the right. The peaks from continuous tubes at  $q \sim 1 \text{\AA}^{-1}$  well reproduce similar peaks from carbon nanotubes of the same diameter with averaging over chiral angle.

The results in Figures 1-4 made it possible to substantially limit the list of nanostructures which may give significant contribution to observed peak at  $q \approx 1 \text{\AA}^{-1}$ . (see Sec. 3)

**3. The local best fits of distribution of carbon nanostructures.** An inverse problem of reconstructing the distribution of carbon nanostructures over a fixed set of structural patterns is formulated as a search for the closest approach from the bottom to the observed curve  $S_{exp}(q)$ , not exceeding it everywhere. The non-unique solutions give the local best fitting, namely, a local minimum, in functional space, of (integrated over  $q$ ) difference of the curves.

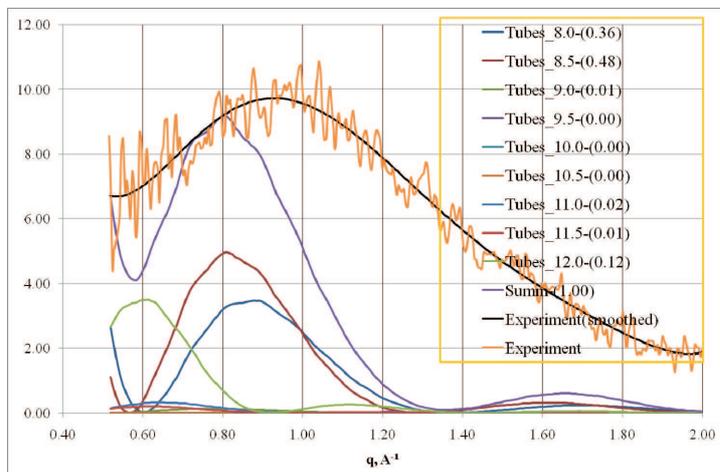


Fig. 5. Local best fitting of observed angular distribution  $S_{exp}(q)$  with the contributions of nanotubes of equal length. Diameter of tube, in  $\text{\AA}$ , and the probability of elementary scatterer (carbon atom) to belong to this structure are indicated.

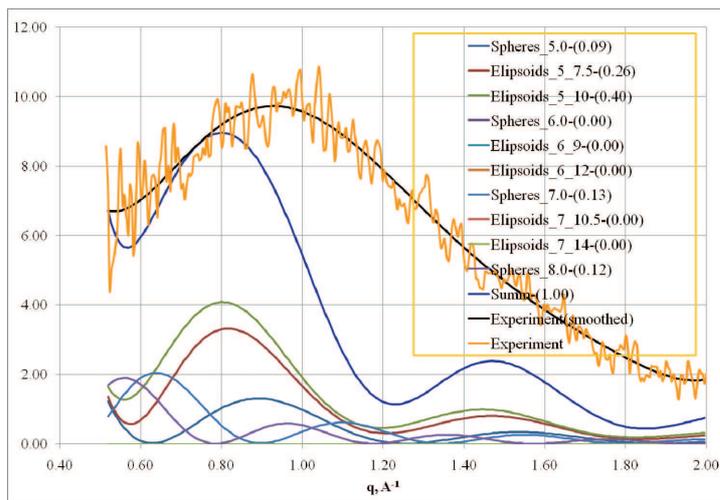


Fig. 6. Local best fitting of observed angular distribution  $S_{exp}(q)$  with the contributions of spheres and ellipsoids. Diameter of spheres, and minor and major diameters of ellipsoids, in  $\text{\AA}$ , and the probability of elementary scatterer (carbon atom) to belong to this structure are indicated.

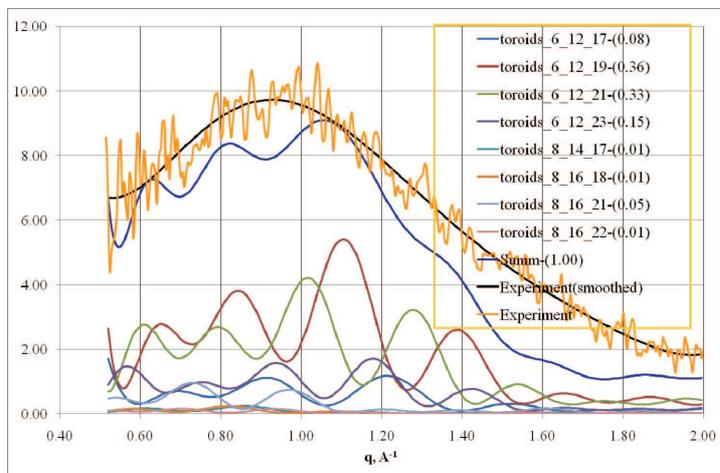


Fig. 7. Local best fitting of observed angular distribution  $S_{exp}(q)$  with the contributions of toroids. Minor and major diameters of elliptic cross-section, and major diameter of toroid, in  $\text{\AA}$ , and the probability of elementary scatterer (carbon atom) to belong to this structure are indicated.

#### 4. Conclusions.

1. The wide peak observed at low scattering angles (scattering wave number  $q \sim 1 \text{ \AA}^{-1}$ ) in the synchrotron-source x-ray diffraction by the films deposited in the vacuum vessel of tokamak T-10 is shown to have a significant contribution of nanostructures composed of single-wall curved graphene sheets. This conclusion is supported by (i) distinctness of the peaks at  $q \sim 3 \text{ \AA}^{-1}$  and  $q \sim 5.5 \text{ \AA}^{-1}$ , which may be explained by nearly universal contribution of randomly-oriented hexagons (and close polygons) in various carbon nanostructures, and (ii) absence of the peaks of quasi-crystal origin (graphite, multiwall nanotubes, fullerite, etc.)

2. For some limited classes of carbon nanostructures, namely (i) nanotubes, (ii) nanospheres and nanoellipsoids, (iii) nanotoroids, an inverse problem of finding the *local* (in a functional space) extremum with respect to approaching (from the bottom) the observed angular distribution in the range of the peak at  $q \sim 1 \text{ \AA}^{-1}$  is solved. The respective probabilities of an elementary scatterer (carbon atom) to belong to the nanostructures of definite topology and size are found.

3. The results suggest the possibility to separate the problems of determination of (a) structural type (namely, topology of single-wall nanostructures with structureless wall), responsible for the observed peak at  $q \sim 1 \text{ \AA}^{-1}$ , and (b) orientational distribution of hexagons (and close polygons) in the above nanostructures from the observed "graphene" peaks at  $q \sim 3 \text{ \AA}^{-1}$  and  $q \sim 5.5 \text{ \AA}^{-1}$ .

4. It may be shown that bigger skeletal structures, assembled from the above-mentioned nanostructures, give minor contribution to the peak at  $q \sim 1 \text{ \AA}^{-1}$ .

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