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Measurement of radio transparency of polycrystalline CVD-diamond in millimeter-wave range by free-space method

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Radio transparency of polycrystalline CVD-diamond disks with diameter up to 75 mm in millimeter-wave range was measured by free-space method. The structure of the disks was characterized by Raman spectroscopy and scanning electron microscopy. Dielectric loss tangent $\tan\delta$ of the samples in the frequency range of 50−67 GHz was found to be in the range of $7.5 \cdot 10^{-3}$−$8 \cdot 10^{-2}$, increasing with frequency. The transmission loss due to the radiation absorption is about 1%.

Keywords: millimeter-waves, polycrystalline diamond, radio transparency.

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Considerable attention to the optical properties of a diamond grown from the vapor phase (chemical vapor deposition, CVD), in the millimeter wavelength range is related to the search for new materials for the output windows of superpower (class 1 MW) continuous-action gyrotrons that generate radiation at the frequency of 170 GHz [1]. There is experimental data on studying the transparency of polycrystalline diamonds at frequencies f up to 1 THz [2–6]. Values of dielectric loss tangent $\tan\delta$ equal to about 10^{-5} [2,3] were measured in the top-quality diamond disks, which corresponds to a low (equal to about $10^{-3}\,\mathrm{cm}^{-1}$) absorption coefficient. As frequency decreases, transparency decreases due to the increasing contribution of free charge carriers to conductivity, the source of such carriers being the diamond defects and boundaries [3]. The frequency domain below 100 GHz is poorly studied and is noted for a large spread of experimental data: the obtained values of $\tan\delta$ in the range of 15-111 GHz are from $\sim 10^{-3}$ to $3 \cdot 10^{-5}$ [4–8]. The frequency range of 50-70 GHz (the five-millimeter wavelength range) is of practical interest given the possibility of high-speed transmission of large information volumes in a broad band, including with the use of 5G mobile communication systems (66-71 GHz). The diamond, having a high thermal conductivity and a wide spectral window of transparency, is a promising material for the making of both radio-transparent radiator covers and heat-conducting enclosures. This paper presents the results of radio transparency measurements for polycrystalline diamonds in the frequency range of 50-67 GHz.

Two polycrystalline diamond disks having the diameter of 57 mm and thickness of 366 μm (sample A) and the diameter of 73 mm and thickness of 450 μm (sample B) were synthesized in the ARDIS-100 Microwave plasma-chemical reactor (Optosistemy LLC, frequency 2.45 GHz) in the methane-hydrogen mixture. The diamond disks were grown on a polished substrate made of single-crystalline silicon 3 mm thick. Sample A was obtained with the following synthesis parameters: methane content in the CH_4-H_2 mixture was 2.5%, pressure 100 Torr, substrate temperature 820°C, deposition rate $\sim 3\,\mu$m/h. Growth conditions for sample B, not transparent in the visible range, were different: CH_4 concentration in the mixture was 5%, pressure 50 Torr, substrate temperature 720°C, deposition rate $\sim 1.5\,\mu$m/h. The total gas flow rate in both processes was equal to 400 sccm. The diamond disk separated from the substrate was obtained by chemical etching of Si in the $HF+HNO_3$ mixture. The images of the growth (large-grained) surface of sample A in the raster electron microscope (REM) (Fig. 1) reveal a typical topography of a polycrystalline diamond having chaotically oriented ground grains. Crystallite size in the sample center is, on the average, $\sim 130\,\mu$m in diameter (Fig. 1, a), but it gradually decreases (as distance from the center increases) to $\sim 50\,\mu$m on the disk edge (Fig. 1, c). Surface texture also changes along the radius, all the more faces (100) appear on the periphery, the number of doubles on faces (111) increases. The value of root-mean-square roughness R_{rms} on the growth side of sample A, measured by means of the NewView 5000 optical profilometer (ZYGO), was in the range of 3-8 μm, decreasing towards the disk edge. Roughness on the opposite smooth side, adjoining the substrate, was about 10 nm. Close values have been obtained for sample B. The assessments have shown that surface roughness must not cause considerable dispersion of Microwave-radiation at the frequencies of 50-67 GHz, that correspond to the wavelength $\lambda \approx 6$−4.5 mm. It is believed that dispersion can be neglected if the ratio $R_{rms}/\lambda < 0.05$.

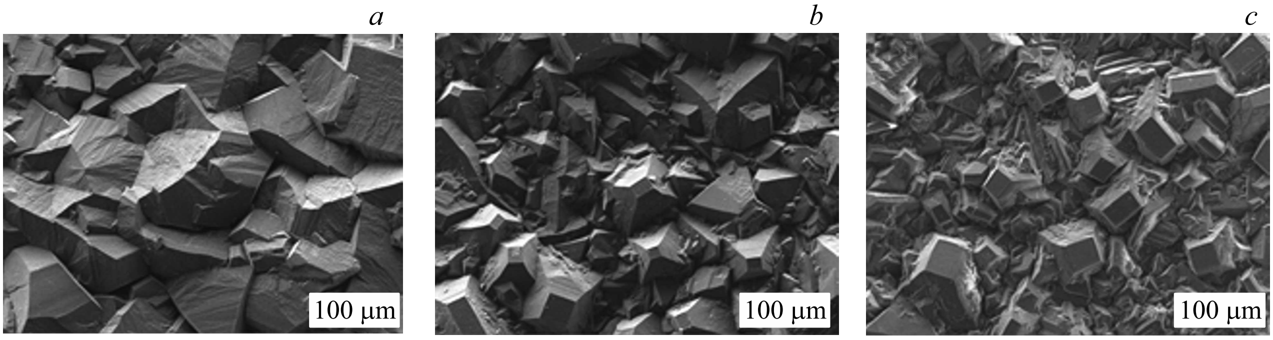

Figure 1. REM-images of the growth surface of a diamond disk 57 mm in diameter (sample *A*): in the disk center (*a*), at the distance of 14 mm from the center (*b*) and near the disk edge (*c*).

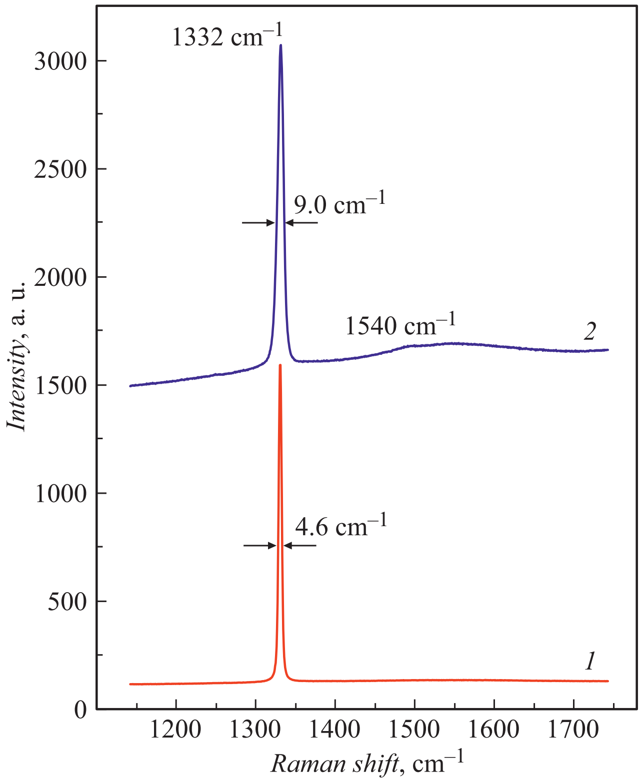

Figure 2. Raman spectra measured in the center of the polycrystalline diamond disk (sample *A*) on the growth side (*1*) and on the substrate side (*2*).

In the given case $R_{rms}/\lambda < 0.0017$, therefore polishing of the disks for absorption measurements was not necessary.

The polycrystalline diamond structure was analyzed by Raman-scattering spectroscopy (RS) of light using the LabRam HR800 spectrometer. Light scattering was excited by a semiconductor laser beam on the wavelength of 473 nm, focused on the sample surface in a spot about 1.5 μm in diameter. Fig. 2 shows the Raman spectra measured on two sides in the disk center: on the large-grained growth side and the diamond side with submicron grains that adjoins the substrate. The growth side has a narrow peak at the frequency of 1332 cm^{-1}, pertaining to the diamond. Other carbon phases are not present in the spectrum, which confirms the rather high quality of the diamond disk. Width of the RS peak in the spectra, recorded along the radius at different distances from the sample center, was within 4.6-6.0 cm^{-1}. A different situation was revealed in the spectra measured on the fine-grained substrate side: in addition to the widened diamond peak 1332 cm^{-1}, a wide weak band with a center at 1540 cm^{-1} is recorded, caused by amorphous carbon inclusions (Fig. 2).

Losses of electromagnetic wave transmission when passing through the diamond disk were measured by the free space method. The method does not impose strict requirements to the geometric shape of the studied sample: it must be only flat with a known thickness within $\lambda/18-\lambda$. To minimize the measurement error due to diffraction effects on the sample edges, sample cross dimension must be equal to at least three widths of the antenna radiation pattern (ARP) by the level of 3 dB in the *E*-plane (in the plane of the electric field vector). The setup layout is shown in Fig. 3, *a*. The disk under study (*1*) was placed in the center between horn antennas with a WR-15 flange (*2*). Distance between antennas was 143 cm, which ensured sample location in the antennas' far zone in the entire frequency range under study. The ARP cross dimension at the distance of 72 cm from the antenna was 10 ± 1.3 cm, exceeding the diamond disk diameter, that's why a radiopaque shield (*3*) with a hole equal to the disk diameter was used. The antennas were connected by means of coaxial-to-waveguide adapters (CWA) and cable assemblies (*4*) to the Keysight vector network analyzer with an operating frequency range of 10 MHz to 67 GHz (*5*).

The value of $\tan\delta$ was determined using the N1500A software application for the Keysight network analyzers for measuring the material properties, which applies the GRL procedure of calibration in free space (gating, reflection, line). The values of ε' and $\tan\delta$ were calculated by the Reflection/Transmission Epsilon Precision (NIST Precision) method based on the measured *S*-parameters. The value of ε' was ~ 5.7. The dependences of $\tan\delta$ on frequency, shown in Fig. 3, *b*, demonstrate a monotonous growth

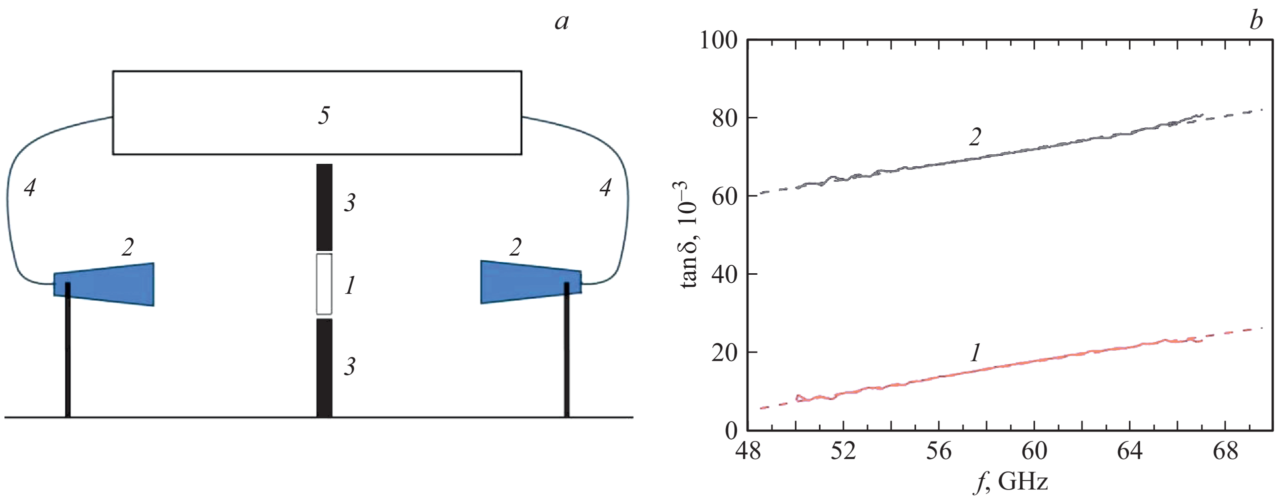

Figure 3. *a* — layout of the experimental setup for Microwave-absorption measurement: *1* — measured sample, *2* — antenna, *3* — shield, *4* — cable assembly with coaxial-to-waveguide adapter, *5* — vector network analyzer; *b* — frequency dependences of loss-angle tangent for diamond disks 57 (*1*) and 73 mm (*2*) in diameter. The dashed lines are approximations using the formula (1).

of losses with frequency. At the frequency of 50 GHz, the value of $\tan\delta$ for disks A and B was $7.5 \cdot 10^{-3}$ and $6.1 \cdot 10^{-2}$ respectively, while at 67 GHz these values increase to $2.2 \cdot 10^{-2}$ and $8.0 \cdot 10^{-2}$. The experimental dependences of $\tan\delta(f)$ can be approximated by expression [3,8]:

$$\tan\delta = a/f + bf, \quad (1)$$

where coefficient $a = \sigma/(2\pi\varepsilon'\varepsilon_0)$, σ is electrical conductivity, ε_0 is electrical permittivity of vacuum, b is constant proportional to concentration of diamond lattice defects. The first equation term describes losses due to conductivity, and the second one — losses caused by single-phonon excitation of acoustic oscillations on lattice defects. The value of $\tan\delta$ in case of high-quality polycrystalline diamonds (partially transparent in the optical range) in the range of 50-200 GHz decreases with frequency approximately proportionally to $1/f$ [3]. Heavily defective diamond disks in paper [8] were characterized by the reverse tendency, namely the increase of losses with frequency rise in the range of 24-30 GHz, which corresponds to domination of the second term in formula (1), the same as in our experiment. The best fit of the measured curves $\tan\delta(f)$ using formula (1) has yielded the following coefficients: $a_1 = -1.20$ GHz and $b_1 = 6.23 \cdot 10^{-4}$ GHz^{-1} for sample A; $a_2 = 0.33$ GHz and $b_2 = 11.10 \cdot 10^{-4}$ GHz^{-1} for sample B.

Mechanisms of occurrence of losses due to conductivity and single-phonon absorption are mainly implemented in the most defective diamond film layer. Amorphous carbon is located chiefly on grain boundaries, therefore the fraction of the amorphous phase for fine-grained diamond films may increase considerably. The primary diamond layer with submicron grains that forms on the silicon substrate is a typical example of such a structure having an increased defect rate. Increase of the defect leads to an increase in the width of the diamond peak at the frequency of 1332 cm^{-1}, which in the spectrum of sample A on the fine-grained (substrate) side reached 9.0 cm^{-1}, while the width of the diamond peak on the large-grained (growth) side is twice less (Fig. 2). Inclusions of electrically-conductive amorphous carbon in a diamond dielectric matrix are usually considered as the main source of Microwave-radiation losses in a polycrystalline diamond [2,3]. A polycrystalline CVD-diamond is a gradient material, and it is the fine-grained substrate layer in the diamond disk that causes, to a large extent, the absorption of optical and IR-radiation [9]. A defect-rich substrate layer can also to a large extent define the value of the second term in equation (1). Removal of the defective layer several tens or even hundreds of micrometers thick by mechanical or laser polishing [9] can facilitate the reduction of Microwave-losses.

The measured minimum values of $\tan\delta = 7.5 \cdot 10^{-3}$ are comparable to the other authors' experimental data for the CVD-diamond (see the Table) that in the frequency range below 70 GHz are within 10^{-3}–$5 \cdot 10^{-5}$. It should be noted that absorption of Microwave-radiation in the diamond depends heavily on structural perfection of the synthesized material and is defined by the growing process technology. Electromagnetic wave absorption coefficient α is related to $\tan\delta$ by ratio $\alpha = (\pi\varepsilon' f \tan\delta)/c$, where f is frequency, while c is speed of light. In the middle of the studied frequency interval (58 GHz), sample A is characterized by the value of $\tan\delta = 1.5 \cdot 10^{-2}$, corresponding to absorption coefficient $\alpha = 0.52$ cm^{-1} and transmission losses 1.3%. Transmission losses of about 1% are acceptable in the use of diamond disks as windows (covers) in designs having a relatively low Microwave-power.

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Literature data on dielectric properties of a polycrystalline CVD-diamond at frequencies below 70 GHz

f, GHz	ε'	$\tan\delta$	Method	Bibliography
15	5.7	$5 \cdot 10^{-4}$	CR	[7]
22	5.7	$1.4 \cdot 10^{-3}$	TE	[5]
24, 30	5.5-5.6	$(1-8) \cdot 10^{-4}$	CR	[8]
27	5.7	$(1.4-5.3) \cdot 10^{-3}$	RR	[4]
36, 72	5.66-5.83	$1.5 \cdot 10^{-4}$, $0.7 \cdot 10^{-4}$	OR	[6]
50	5.7	$7.5 \cdot 10^{-3}$	FS	This work
60	-	$5 \cdot 10^{-5}$	FP	[3]

Note. CR is cylindrical resonator; TE is resonator with TE mode; RR is reflecting resonator; OR is open resonator; FS is free space method; FP is Fabry-Perot resonator.

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Conflict of interest

The authors declare that they have no conflict of interest.

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