Terahertz Conductivity of Plasma-treated of CNT-based Macroscale Films

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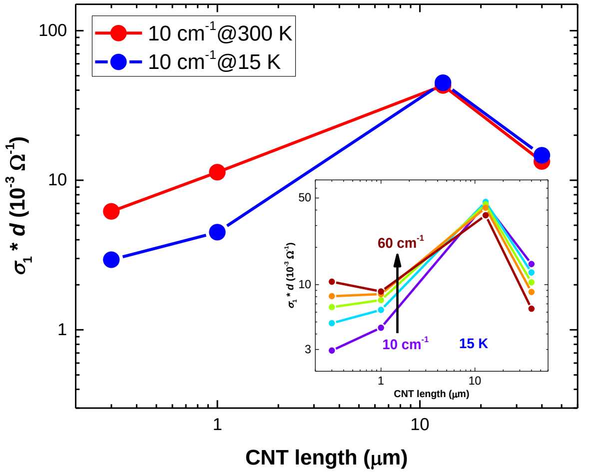
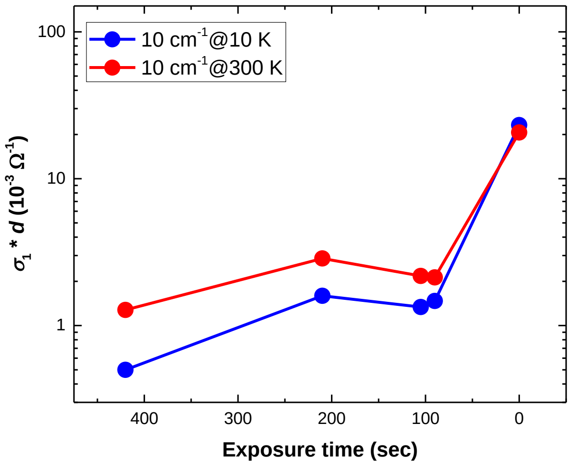
Time-domain terahertz and Fourier-transform infrared spectroscopy techniques are used to study temperature-dependent conductivity spectra of free-standing macroscale films composed of disordered single-walled carbon nanotubes. Films composed of nanotubes a) treated with oxygen plasma and b) having different lengths are used.

We observed evolution of the so-called terahertz conductivity peak during the change of the temperature and the length of nanotubes. We analyzed changes of the terahertz conductivity of the films from metal-like to semiconductor-like due to oxygen plasma treatment and shortening length of the nanotubes.



Dependence of the conductance of films at the frequency 10 cm^{-1} on the length of CNT



Dependence of the conductance of films at the frequency 10 cm^{-1} on plasma treatment time

References

[1] B.P. Gorshunov, E.S. Zhukova, Ju.S. Starovatykh et al., *Carbon* V.126, 544-551 (2018)
[2] D.S. Kopylova et al., *Nanoscale* vol.10, 18665-18671, (2018)
[3] E.S. Zhukova, B.P. Gorshunov, A.P. Tsapenko et al., *Journal of Physics: Conference Series*, 1092, art. no. 012178 (2018)
[4] E.S. Zhukova, A.K. Grebenko, A.V. Bubis et al., *Nanotechnology* , V.28, N.44 (2017)
[5] A. Kaskela, A.G. Nasibulin, M.Y. Timmermanset al., *Nano Lett.* 10, 4349–4355 (2010)

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The macroscale films of disordered high-quality single walled carbon nanotubes (SWCNT) were synthesized by an aerosol CVD (floating catalyst) method. Two types of the macroscale films were studied: a) composed of CNTs treated by oxygen plasma [2] with exposure time of 1,5, 10, 30, 90, 105, 180, 210 and 420 seconds and b) composed of high-quality pristine SWCNTs [5] of average lengths of CNTs of 0.3, 1, 6, 13 and 40 μm. In the experiments, free-standing films and films on the 5 μm thick polyethylene substrate were used.

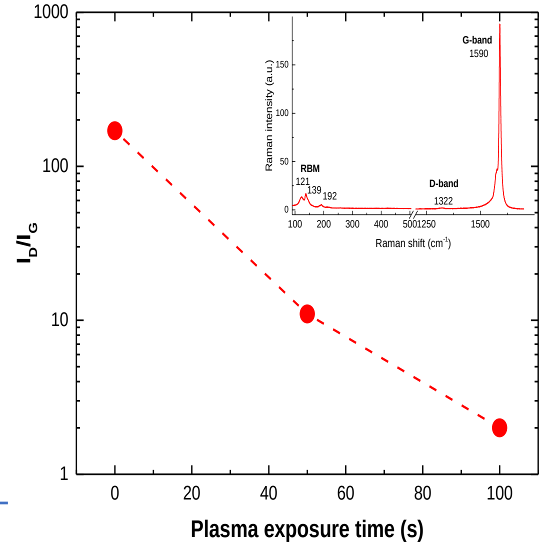

Fig.2 Dependence of the I_G/I_D ratio on the exposure time, on the inset Raman spectra of pristine [2]

Oxygen plasma treatment:

- Cuts the tubes to a shorter parts
- Defects of the tube's wall
- Doping
- Increase the amount of carboxyl group

Thickness of the Films with different length of CNT

Length, μm	0,3	1	6	13	40
Thickness, nm	380	~300	~300	340	50

Fig.1 CNT film on the aperture (aperture diameter 5 - 8 mm)

Due to the friability of the films, their thickness *d* cannot be accurately determined. Therefore, all the results were analyzed in terms of complex conductance:

$$\textbf{Conductance} = \sigma(\nu)\cdot d$$

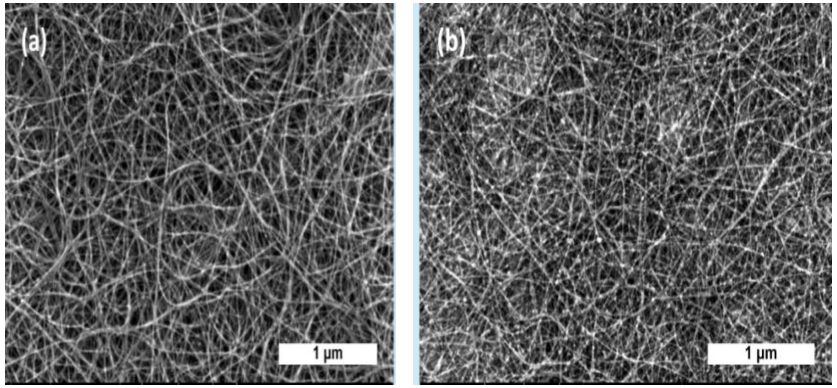

Fig.3 SEM images of (a) pristine SWCNT film placed onto quartz substrate, (b) the same film treated with oxygen plasma with the treatment time of 100 s [2].

[2] D.S. Kopylova et al., *Nanoscale* vol.10, 18665-18671, (2018)

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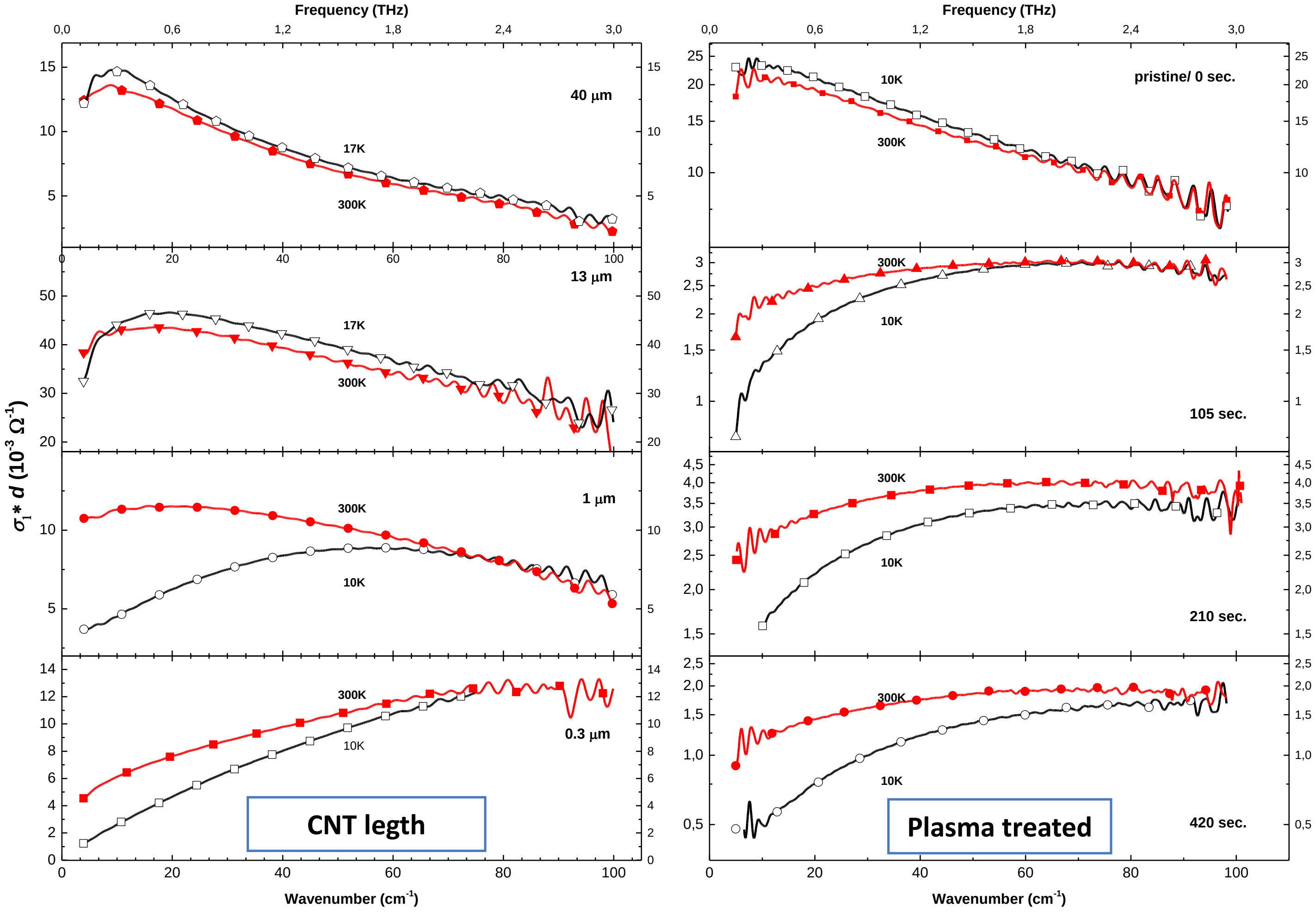
Fig. 4 Real part of conductance, $\sigma_1(\nu)*d$, of films composed of CNT with average lengths of 0.3, 1, 13 and 40 µm (left panel) and films treated with oxygen plasma with an exposure of 0, 105, 210 and 420 seconds (right panel). Red symbol/ine correspond to the measurement at room temperature, black – at 10 K - 17 K



Experimental details

Infrared Fourier-transform spectrometer Vertex80v and pulsed terahertz time-domain spectrometer TPS spectra 3000 were used for measurements in the 5 to 15000 cm-1 frequency range. The spectra of real σ_1 and imaginary σ_2 parts of the complex conductivity for the 5 to 100 cm-1 frequency range were directly determined from the time-domain data. Infrared (IR) conductivity spectra were obtained by modeling the IR transmission coefficient data with the Drude-Lorentz approach and merged with directly measured THz data to obtain broadband (5 cm-1 to 15000 cm-1) spectral response of the films. Homemade gas-flow optical cryostat was used to examine the films in the temperature range from 4 to 300 K.

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Introduction | Objects | Results | Conclusion

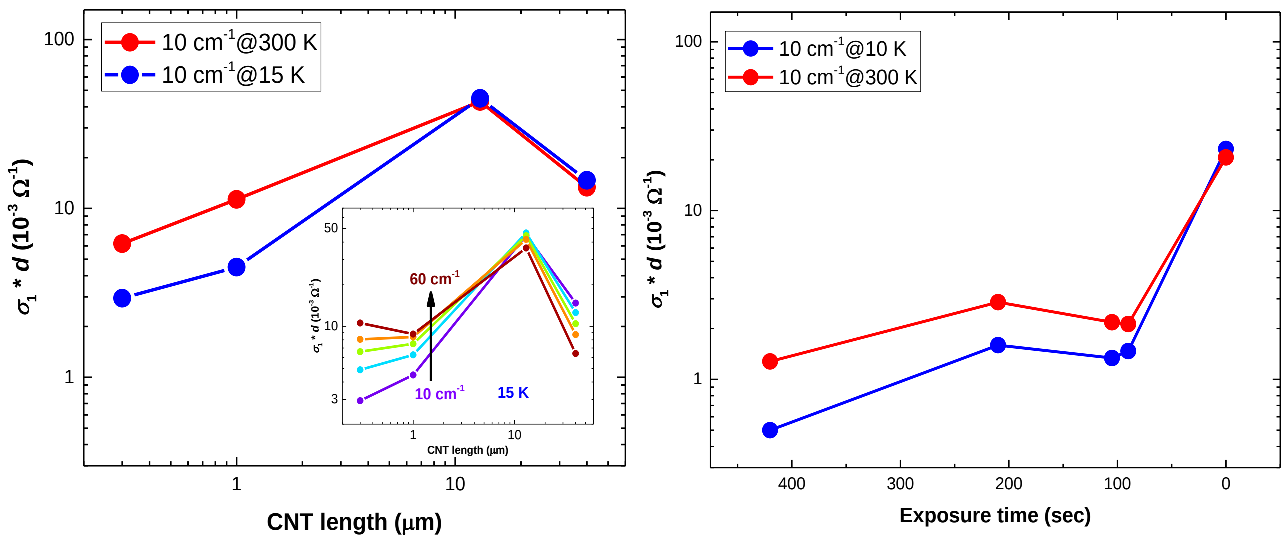

Fig. 5 Real part of conductance $\sigma_1(\nu)*d$ at a frequency 10 cm^{-1} of the films composed of CNTs with average lengths of 0.3, 1,13 and 40 µm (left panel), and of pristine CNTs (0 seconds) and treated with oxygen plasma films with an exposure time of 105, 210 and 420 seconds (right panel). Red symbols/lines correspond to the measurements at room temperature, blue – at 15 – 17 K. On the Inset on the left panel length dependence of the real part of conductance at frequencies in the range 10 – 60 cm^{-1}.

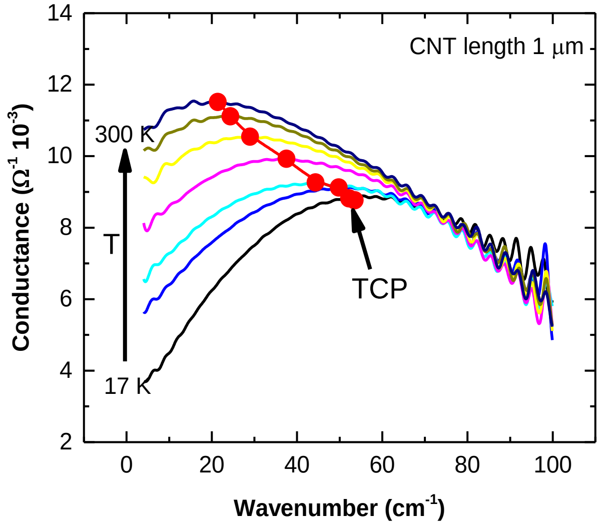

Fig. 6 Real part of conductance $\sigma_1(\nu)*d$ of the films composed of 1 µm long CNTs. Red points mark the terahertz conductivity peak position for temperatures from 17 K to 300 K.

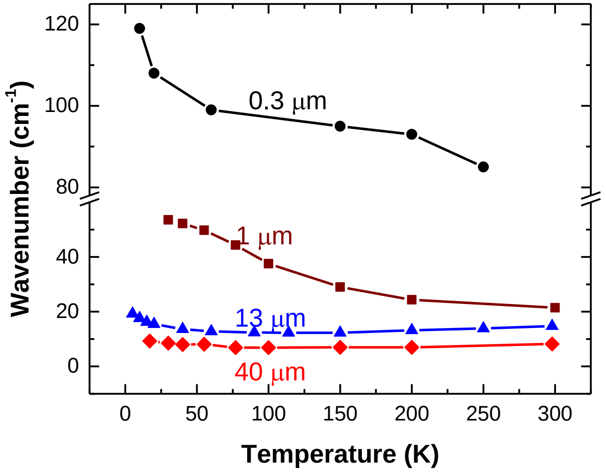

Fig. 7 Temperature dependence of the THz conductivity peak position for the films composed of CNTs with average lengths of 0.3, 1, 13 and 40 µm. Points for film with 0.3 µm CNTs are obtained by modeling with the Drude-Lorentz model.

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1) Character of the temperature dependence of the conductance of the samples with long CNTs, 13 and 40 µm, exhibits metal-like temperature behavior. This means that in such films the main scattering mechanism for charge carriers is determined by phonons.

2) For the plasma treated films and films composed of short CNTs (0.3 and 1 µm) this dependence becomes progressively more semiconductor-like with the exposure time and with shortening the CNTs length.

Taking into account that oxygen plasma exposure leads to appearance of defects in individual tubes [2], we assume that the main contribution to the temperature dependence of films conductance comes from the tunneling of charge carriers through the inter-tube contacts and defects in individual CNTs, since with decreasing the temperature the kinetic energy of charge carriers becomes smaller.

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