Dielectric properties of vertically aligned carbon nanotubes in the mid-IR and THz spectral range

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Vertically aligned carbon nanotubes (VACNT) are interesting candidates as chemo-physical sensors. The reason lies not only in their huge surface-to-volume ratio, but also the low carrier dimensionality which suggests that an adsorption of molecules should lead to a measureable change of the conductivity. Here, we explore the use of THz radiation to probe for such conductivity changes in a contactless manner.

In preparation of sensing measurements, we first investigated the dielectric properties of VACNT of different tube lengths in the THz/mid-IR spectral range (0.1-100 THz) [1], because the data found in the literature are highly disparate and their interpretation is conflicting. Our measurements with the radiation impinging vertically onto the sample yield a rather featureless optical density with a broad peak around 24 THz.

We employ for the first time a fit of such data with a *Bergman*-type effective medium theory which takes the morphology of the tubes into account via depolarization factors which can be understood as representing the capacitive coupling between neighboring tubes respectively the internal capacitance of each tube. The intrinsic conductivity of the tubes is assumed to be of conventional Drude type. With this model, we can fit our measure data very satisfactorily if we allow for a weak additional axial conductance manifesting at low THz frequency and arising because the tubes are not perfectly vertical. The fits yield a plasma frequency $\nu_p = 42\pm2$ THz, a carrier momentum scattering time $\tau = 3.5\pm0.3$ fs and a residual axial conductivity of ~0.3 $(\Omega\text{cm})^{-1}$. Our analysis suggests that the responses of plasmonic, phononic, Drude-Smith, Drude-Lorentz or even more exotic type resorted to in the literature in order to explain the THz or IR spectra may in most cases not be needed and may only mimic the depolarization effects imposed by the nanostructure of the tubes and their microscopic arrangements.

We also show first THz gas sensing measurements which reveal doping effects of NH_3 and SO_2 adsorbed in the VACNT "forest" [2]. Fig. 1 displays results from our THz transmission measurements of VACNT in either air or NH_3 atmosphere. The presence of NH_3 reduces the conductivity. This can be understood – consistent with the literature – by the donor character of the NH_3 molecules. VACNT are a statistical mixture of metallic and semiconducting tubes with an overall p-type conductivity. The interaction with the NH_3 molecules reduces the hole density and with it the conductivity. This suggests that THz read-out of VACNT may be useful for gas-sensing.

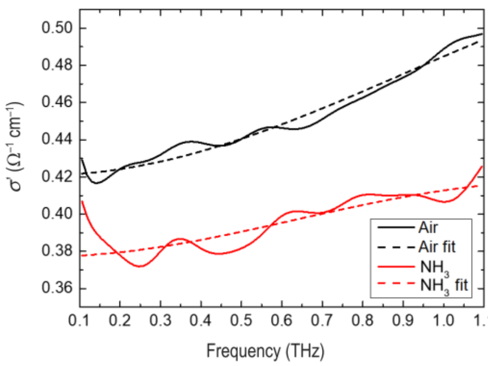

Fig.1 Decrease of the THz conductivity of VACNT upon exposure to NH_3 gas at ambient pressure. The upper curve (black online) is for VACNT in air, the lower one (red online) for VACNT in NH_3 atmosphere.

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[1] M. D. Thomson *et al.*, J. Phys. D: Appl. Phys. **51**, 034004 (2018).
[2] W. Zouaghi *et al.*, Lith. J. Phys. **58** (2018) in press.