Terahertz analysis reveals nanostructural dependence of carbon thin-film properties

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When thin films made from different types of carbon nanotubes are exposed to electromagnetic radiation in the range of 0.1–2THz, the response reveals the effects of surface defects on the optical and electrical properties.

Terahertz (THz) technology has great potential to improve nanoscience-based devices. The high-performance optoelectronic setups that generate and detect terahertz electromagnetic signals could extend the capabilities of a range of applications, including sensing, imaging, and telecommunication. Analysis and signal processing using coherent terahertz time-domain spectroscopy (THz-TDS) provide a precise and simple approach to measuring the response of a material to electromagnetic absorption and dispersion over the frequency range 0.1–2THz. For example, from the THz-TDS measurements of the flat parallel faces of a sample film we can derive both the refractive index and the power absorption coefficient of different materials.

Graphene (an atomically thin honeycomb lattice of carbon) and carbon nanotubes (CNTs; rolled up tubes of graphene) are materials at the forefront of state-of-the-art nanotechnology research in one, two, and three dimensions. A highly sensitive response to changes in the environment has been demonstrated for the properties of CNTs compared with conventional structures. Their high sensitivity makes CNTs ideal candidate materials in a range of industries, such as biomedicine and security. The high mobility and electron transfer at terahertz frequencies also lend them to applications based on photonic emission.

Graphene carriers quickly relax to equilibrium following excitation, with a relaxation time of less than 1ps. The rate of recombination of electrons and holes excited in graphene is slow—taking times greater than 1ps. In addition, graphene has a gapless energy spectrum in the terahertz range, so it is a perfect conductor at these frequencies. As a result, graphene has stimulated research in a wide range of applications, and it has been used for the study of intraband and interband electronic processes through observations of optically pumped amplified stimulated terahertz emission.

CNT thin-films comprise both metallic and semiconducting CNTs embedded in air. Networks of CNTs can form in the films, which can be used in metal–semiconductor, semiconductor–semiconductor or metal–metal junctions. In this work, we have used two thin films, one comprising single-walled CNTs (SWNTs) and the other multiwalled CNTs (MWNTs) on a transparent fused quartz substrate (see Figure 1). The power absorption of the substrate is less than 10cm⁻¹ at 2THz. Deposition of the carbon nanotube thin-film on a gate insulator with a high dielectric permittivity, such as quartz, has the advantage that it does not degrade the perfect high-speed carrier transport of the CNTs.

We performed the terahertz characterization of the CNT sample using a classical THz-TDS setup: see Figure 2(a). We made three measurements of temporal waveforms: see Figure 2(b). The first measurement recorded the reference signal without a sample in between the antennas. The second measured the signal with the quartz substrate in the setup, and the third measured the signal with the substrate covered by the CNT film.

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At higher frequencies, the measured power absorption of the SWNT thin film is higher than the MWNT film: see Figure 3(a). This can be attributed to the larger length and diameter of MWNTs (which are 15μm long and 15nm in diameter) compared with SWNTs (5μm long and 1.5nm in diameter). The order of magnitude of the absorption and refractive index of both MWNTs and SWNTs (see Figure 3) is about 10 times greater than those reported by Jeon et al.8 This is most likely due to the higher CNT density of our CNT films than in the samples studied by Jeon. The frequency response curves for both refractive index and absorption are those of a metallic layer with a plasma frequency at several terahertz.

Figure 4 shows our recent experimental results for a THz-TDS analysis of the real part of the complex conductivity—that is, the optical response—for the measured absorption and index of refraction as in Figure 3. Importantly, the SWNT sample has a greater conductivity compared with the MWNT film because of the low level of surface defects in SWNTs. Jung et al.9 have compared the real part of the conductivity for different carbon nanostructures. Their results show that the real part of the conductivity is strongly dependent on the crystallinity of the graphitic layers and the number of surface defects in the carbon nanostructure. As a result, the value of the power absorption, refractive index, and conductivity is greater for double-walled carbon nanotubes compared with MWNTs and still greater again for SWNTs.

In summary, we have studied the terahertz frequency optical conductivity response of SWNT and MWNT thin films. Using THz-TDS, we have observed that the power absorption coefficient, reflective index, and conductivity of a SWNT
Figure 4. The real part of the conductivity of the single-walled and multiwalled carbon-nanotube (SWNT and MWNT) thin films.

The real part of the conductivity of the single-walled and multiwalled carbon-nanotube (SWNT and MWNT) thin films. The sample is much higher than for a MWNT sample because of the higher number of surface defects in MWNTs.

The future work of our research group will focus on studying the THz-TDS of composite films of CNTs and silver nanoparticles. The work will be carried out in collaboration with a Korean research group in order to find the effect of atomic interaction on the number of surface defects and, consequently, the possibility of increasing the conductivity of MWNT samples for further nanoscience applications.

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References

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