Ultrafast changes in the far-infrared conductivity of carbon nanotubes

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***Abstract*—The ultrafast charge-carrier dynamics in single-wall carbon nanotubes (NTs) have been investigated by time-resolved THz spectroscopy. Both the equilibrium and non-equilibrium conductivity data of the NTs in the far-infrared (FIR) spectral range from 1 to 40 THz are dominated by optical transitions across the band gap of tubes with gap energies of ~ 10 meV. A simple model based on an ensemble of two-level systems excellently explains all experimental findings. In particular, the surprisingly weak temperature dependence of the FIR conductivity has been shown to arise from tube-to-tube variation of the chemical potential which is ~ 100 meV in our sample. The results strongly suggest to use the temperature dependence of the FIR conductivity as a very sensitive and contact-free probe of the NT sample purity. Finally, the relaxation of the photo-excited NT sheet on a picosecond time scale mainly reflects the cooling of hot phonons which is about five times faster than in graphite. This points to much stronger lattice anharmonicities in NTs.**

I. Introduction And Background

Single-wall carbon nanotubes consist of rolled-up hexagonal sheets of carbon atoms where the tube diameter and chirality determine whether the tube is semiconducting or purely metallic[1]. The energy gap of semiconducting tubes with a diameter around 1 nm lies in the 10-meV or 1-eV range for small- and large-gap tubes, respectively. This broad variety, the small diameter, and the high threshold for electronic breakdown make NTs unique and promising elements of future nanoelectronics. In order to realize such potential, a comprehensive knowledge of the charge carrier dynamics in NTs is essential. Time-resolved THz spectroscopy is a promising experimental approach to this task. A visible pump pulse excites charge carriers in the sample and a subsequent THz pulse probes the low-energy response of the system such as transport relaxation of charge carrier near the Fermi level.

II. Experiment and Results

The experimental setup including the Ti:sapphire-laser based THz spectrometer for static and time-resolved measurements in the 1 to 40 THz spectral range together with the sample preparation are given in detail in Refs.[2-4]. In large-gap tubes (typically samples contain NTs of all chiralities, i.e. semiconducting and metallic species), the lack of a free carrier response in our time-resolved data is consistent with the immediate generation of strongly bound excitons with binding energies exceeding our THz probe energies by far[2] (see Fig. 1a). The optical properties of such NT samples are crucially determined by interband transitions across the energy gap of small-gap species[2-5] (see Fig. 1b). The tube-to-tube variation of the chemical potential with respect to the gap center naturally explains the weak temperature dependence of the FIR absorption which has been controversially discussed in the literature. Our model is also capable to reproduce the instantaneous conductivity of a NT sample that has been pumped by a short visible laser pulse before arrival of the THz probe pulse. In this way, the energy relaxation of the optically excited electrons is monitored. We finally argue that the pump-excited electrons heat up only a few strongly coupled phonon modes. The decay of the pump-induced signal mainly reflects the cooling of these hot phonons and is substantially faster than in graphite[5] pointing to stronger lattice anharmonicities.

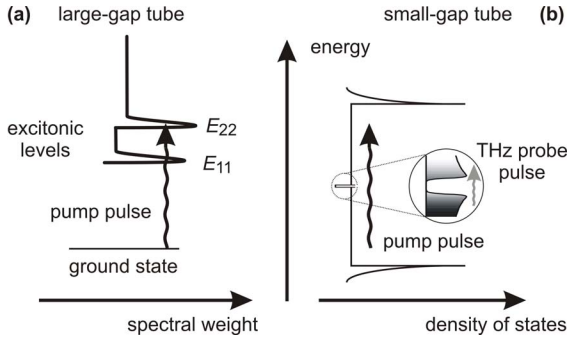

Fig. 1: Excitonic levels of a large-gap NT together with a visible pump pulse resonantly exciting the E_{22} band. (b) Single-electron density of states of a small-gap NT with a magnification of the small band gap. Wiggled arrows mark transitions induced by the visible pump and the THz probe photons, respectively. The various gray scales represent different electronic occupation numbers.

References

[1] *Carbon nanotubes: Synthesis, structure, properties, and applications*, M. S. Dresselhaus, G. Dresselhaus, and P. Avouris, Eds. Berlin: Springer, 2001.

[2] L. Perfetti, T. Kampfrath, F. Schapper, A. Hagen, T. Hertel, C. M. Aguirre, P. Desjardins, R. Martel, C. Frischkorn, and M. Wolf, "Ultrafast dynamics of delocalized and localized electrons in carbon nanotubes", Phys. Rev. Lett. **96**, 027401 (2006).

[3] T. Kampfrath, L. Perfetti, K. von Volkmann, C. M. Aguirre, P. Desjardins, R. Martel, C. Frischkorn, and M. Wolf, "Optical response of single-wall carbon nanotube sheets in the far-infrared spectral range from 1 THz to 40 THz", phys. stat. sol. (b) **244**, 3950 (2007).

[4] T. Kampfrath, K. von Volkmann, C. M. Aguirre, P. Desjardins, R. Martel, M. Krenz, C. Frischkorn, M. Wolf, and L. Perfetti, "Far-infrared conductivity of carbon-nanotubes films", Phys. Rev. Lett. (submitted).

[5] T. Kampfrath, L. Perfetti, F. Schapper, C. Frischkorn, and M. Wolf, "Strongly coupled optical phonons in the ultrafast dynamics of the electronic energy and current relaxation in graphite", Phys. Rev. Lett. **95**, 187403 (2005).

978-1-4244-2120-6/08/$25.00 ©IEEE.