Terahertz Driven Opacity-Transparency Transition in Photoexcited Carbon Nanotubes

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Abstract: CNTs exhibit extraordinary nonlinear THz responses upon optical excitation. Its conductivity reduces at intermediate intensities, while soaring elsewhere. Field-effect mobility and carrier multiplications govern the rise and fall of the conductivity.

High-field electron dynamics in carbon nanotubes (CNTs) is of great interest considering its potential for high-speed nanoelectonics applications.[1] We have demonstrated that strong THz pulses induce nonlinear absorption in free-standing CNTs.[2] The field-induced absorption monotonically increases as the field gets stronger (gray line in Fig.1a). Intense THz fields release carriers from localized states and generate carriers via interband tunneling and impact ionization, and hence transiently enhance the permittivity of the CNTs. When the CNTs are optically excited, however, the CNTs exhibit drastically different THz responses. THz transmission of the photoexcited CNTs is not monotonically decreasing as the field strength increases. It rises in the intermediate range of the THz field strength between 540 and 660 kV/cm (blue line in Fig.1a), while falling below and above the region. We investigate the field induced opacity-transparency transition in photoexcited CNTs with high-field THz spectroscopy.

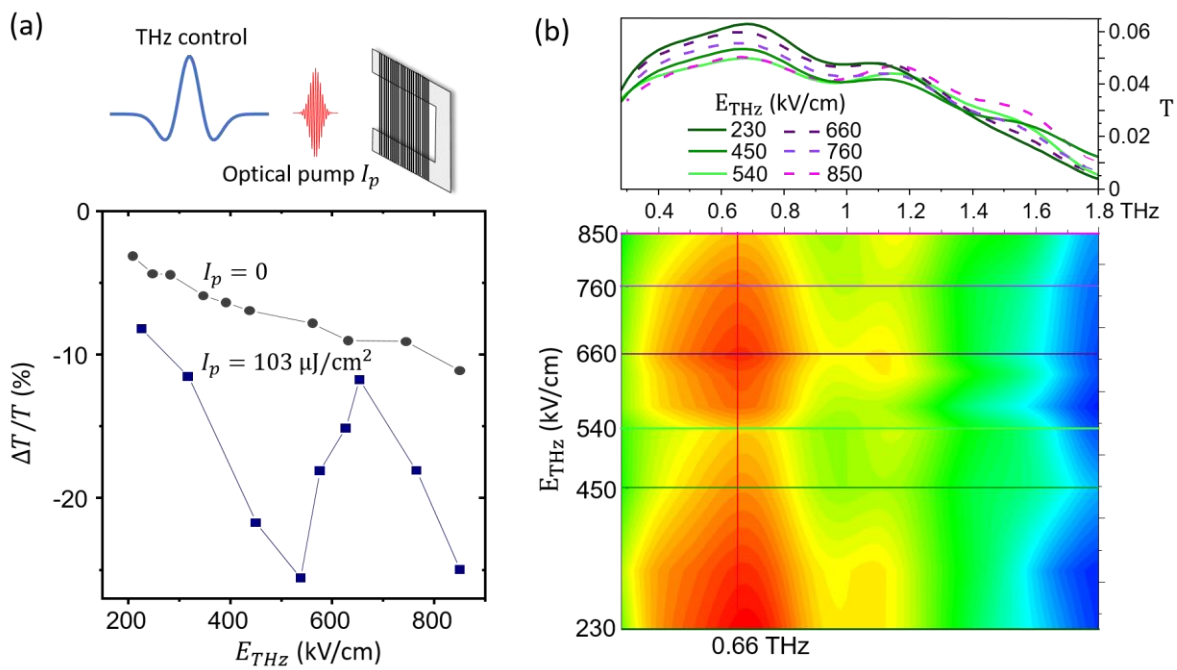

Fig. 1. (a) High-field induced nonlinear THz transmission of CNTs with and without optical excitation (b) THz transmission spectra of the photoexcited CNTs in the range of the incident peak field, $230 < E_{THz} < 850$ kV/cm, when the optical pump fluence is 103 μJ/cm^2. Top panel: transmission spectra at $E_{THz} = 230, 450, 540, 660, 760, 850$ kV/cm

We conducted time-resolved high-field THz spectroscopy of optically excited free-standing CNTs.[2] We drew CNT sheets (thickness, 50 nm) from a forest of multi-walled CNTs (diameter, 10 nm; averaging 9 walls per tube) and wound them on a U-shaped polyethylene (PE) reel (thickness, 125 μm). THz transmission for the polarization parallel to the CNT axis is 8.9%. Strong THz pulses were generated by optical rectification of femtosecond laser pulses (pulse energy, 1 mJ; pulse duration, 100 fs; repetition rate, 1 kHz) with titled pulse front for phase matching between optical and THz pulses in $LiNbO_3$.[3] The field amplitude of the broadband THz pulses (central frequency, 0.7 THz; bandwidth, 0.7 THz) reached 0.9 MV/cm. We detected THz pulses using a liquid-He cooled Si:Bolometer and traced the waveforms with electro-optic (EO) sampling with a 1-mm ZnTe crystal. Figure 1b presents the spectra of the THz pulses transmitted through the photoexcited CNTs depending on the incident THz field strength. The contour plot clearly shows that the fields enhance transparency in the intermediate intensity range between 540 and 660 kV/cm, while raising opacity below and above the region.

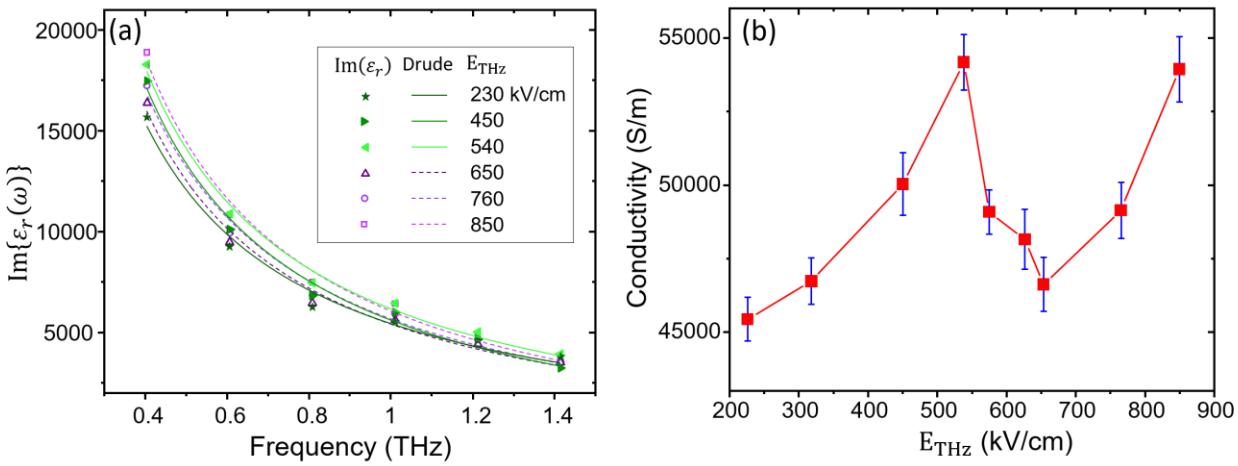

Fig. 2. (a) Imaginary part of the relative permittivity of the photoexcited CNTs for E_{THz} = 230, 450, 540, 660, 760, 850 kV/cm. The solid and dashed lines are fits to the Drude model. (b) Conductivity versus peak field amplitude

We analyzed the amplitude and phase spectra of the transmitted waveforms to extract the complex refractive index of the CNTs using the Fresnel transmission coefficient. The solid green and open violet symbols shown in Fig. 2a are the imaginary part of the relative permittivity, $\varepsilon_r(\omega)$, of the photoexcited CNTs for E_{THz} = 230, 450, 540, 660, 760, 850 kV/cm. We fit the extracted relative permittivity with the Drude model

$$\varepsilon_r(\omega) = \varepsilon_b + i\frac{\sigma_0}{\varepsilon_0\omega} \quad (1)$$

where σ_0 is the conductivity and ε_b is the bound-electron permittivity. The solid green and dashed violet lines in Fig.2a present the fitted curves, from which we obtain the conductivity of the photoexcited CNTs. Figure 2b shows the conductivity as a function of the peak field amplitude, confirming the suppression of conductivity in the region of intermediate intensity.

We speculate that the competing processes, field-effect mobility and field-induced carrier multiplications, govern the rise and fall of the conductivity. Releasing charge carriers from localized states, strong THz fields increase carrier mobility in CNTs, and, consequently, raise conductivity. The initial rise of the conductivity up to 540 kV/cm is accounted for by the mobility increase of both the photoexcited hot carriers and the cold carriers in the conduction band. When the THz fields exceed 540 kV/cm, most of the hot carriers are freed from the localized states, and field induced scattering such as side band scattering and carrier-carrier scattering become dominant high-field effects. The field induced scattering lowers the carrier mobility and decreases the conductivity. Another possible mechanism of the conductivity decrease is field-enhanced charge recombination, which has been observed in some organic semiconductors.[4] In the high field regime above 660 kV/cm, carrier multiplications via impact ionization and interband tunneling are substantial to increase the conductivity. Clarifying the microscopic processes awaits future investigation.

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References

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