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High-field terahertz time-domain spectroscopy of single-walled carbon nanotubes

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Abstract. We experimentally demonstrated the high-field THz response of Single-Walled Carbon Nanotubes (SWCNT) in the broad frequency range from 0.2 to 2.0 THz. To investigate the impact of nanomaterial geometry on the absorption of THz radiation, two kinds of SWCNT films with different diameters and lengths were fabricated. The measured conductivity shows the change that can be attributed to the change of the Drude term of conductivity. This increase in conductivity at lower frequency was described either as the increase in the number or decrease in effective masses of free charge carriers different for two samples. Our study suggests that the conductivity of the SWNTs in strong THz fields is enhanced by inducing strong non-linear electron dynamics as a result of several competing processes. Our findings can be used to predict the behavior of CNT devices (modulators, polarizes, lenses, etc.) in the THz high-field.

Keywords: carbon nanotubes, high-field THz spectroscopy, THz spectroscopy

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Спектроскопия временного разрешения одностенных углеродных нанотрубок в высоких терагерцевых полях

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Аннотация. Мы экспериментально исследовали ТГц-пропускание одностенных углеродных нанотрубок (ОУНТ) в широком диапазоне частот от 0,2 до 2,5 ТГц при различных мощностях ТГц поля. Для исследования влияния геометрии наноматериала на поглощение терагерцового излучения были изготовлены два вида пленок, содержащих ОУНТ разного диаметра и длины. Измеренная проводимость для двух образцов можно объяснить изменением Друде-части проводимости. Это увеличение на более низкой частоте можно объяснить либо увеличением числа, либо уменьшением эффективных масс свободных носителей заряда, различных для двух образцов. Наше исследование предполагает, что в сильных терагерцовых полях проводимость ОСНТ увеличивается за счет создания сильной нелинейной динамики электронов в результате нескольких конкурирующих процессов. Результаты этой работы могут быть использованы для прогнозирования поведения устройств на основе УНТ (модуляторов, поляризаторов, линз и т. д.) в высоких терагерцовых полях.

Ключевые слова: углеродные нанотрубки, мощные терагерцовые поля, терагерцовая спектроскопия

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Introduction

It is well-established that carbon nanotubes (CNT) exhibit unique properties associated with their quasi-one-dimensional structure, such as 1D ballistic conduction, large exciton binding energies, and strong many-body interactions. A thin-film CNT network shows exceptionally high effective macroscopic THz conductivity described by the Drude and plasmon models, where some fraction of free charges undergoes plasmon resonance, representing the confined collective motion of carriers along tubes, while other charges undergo a Drude-like free carrier response, indicative of delocalized intertube transport [1]. Numerous THz devices have been proposed based on these properties [1]. On the other hand, the influence of CNT response at internal electric fields up to 130 kV/m, important for THz communications, has been rarely studied.

A particular experimental study has found that strong THz fields generate excitons in semiconductor single-walled CNTs [2]. The free standing multi-walled CNT (MWCNTs) in intense THz field showed anisotropy both in linear and in nonlinear effects [3]. Oppositely to a previously reported article [2], theoretical analysis based on the Drude model suggests that strong THz fields enhance the permittivity of the MWNTs. In recent research [4], extraordinary nonlinear terahertz responses upon optical excitation were observed. It was attributed to field-effect mobility and field-induced carrier multiplications, which were considered to be competing processes governing the rise and fall of the conductivity.

In this study, we present an experimental investigation of the high-field THz time domain spectroscopy of two SWCNTs samples with different geometries. The strong THz radiation results in the non-linear absorption of SWCNTs, which is opposite to the field-induced transparency of SWCNTs typically observed for conducting medium. It was attributed to an increase in the number of free charge carriers accompanied by the decrease in the effective masses, which saturate at high field strengths.

Materials and Methods

SWCNT films with diameters ranging from 1.2–1.7 nm and length from 300 nm to 1 μm, and 1.6–2 nm with the corresponding lengths from 1 to 5 μm were studied, similarly to those published in [5]. The average thickness of all films was less than 100 nm. To investigate the impact of geometry (diameter and length) on the THz conductivity of free carriers at high field strengths, thin films of materials were produced by spray coating on z-cut quartz substrates.

Terahertz time-domain spectroscopy (THz-TDS) of SWCNTs was measured on the experimental setup based on the femtosecond Yb-doped diode-pumped solid-state amplifier system with a wavelength of 1030 nm and pulse duration of 30 fs and repetition rate of 1 kHz. For THz radiation emission, the BNA organic crystal was used. The generated THz radiation power was about 520 uW (measured by Ophir THz power meter RM9) and focused on CNT by the off-axis parabolic mirror to the spot of about 900 μm diameter. The duration of a THz pulse was about 1.25 ps, and its spectrum was up to 2.5 THz wide. The THz electric field strength was up to 130 kV/cm. The ZnTe crystal of 1mm thickness was used as a detector. The absence of nonlinear effects in the detection of a signal on a ZnTe crystal can be indirectly verified by checking the monotonicity of the dependence of the transmission and related parameters on the THz field. In addition, it is known from the literature that nonlinear effect appears at power significantly exceed 1 MV/cm [6].

Results and Discussion

Fig. 1, *a* shows the change the sheet conductivity for shorter SWCNT. The equilibrium conductivity of SWCNT films is well described by the Drude-plus-plasmon model [1]. With the increase in field strength, the conductivity increased in the low frequency range, indicating the change in the Drude component. We then used a model-independent approach, as described in [7], to evaluate the ratio of the effective density of electrons to effective mass, N_{eff}/m (Fig. 1, *b*). The magnitudes of the real part of the sheet conductivity at lower frequencies show the general tendency, increasing as the field strength goes up (Fig. 1, *c*). There are two main competing processes that are found to be influenced by the observed conductivity behavior [3, 4]. Firstly, intense THz fields reduce the conductivity of metallic materials, as an example of graphene, by increasing the electron temperature accompanied by the decrease of the scattering time [8]. On the other hand, conductivity goes up if intense THz fields increase carrier density by generating carriers by carrier multiplication by impact ionization and field induced interband tunneling [2]. These competing processes might be reflected in N_{eff}/m as follows the increase of the carrier density will result in the increase of N_{eff}/m, while the subband scatterings give rise to a higher effective mass for the carriers N_{eff}/m. Therefore, we calculated the ratio N_{eff}/m to evaluate this observation. In our experiments, both samples showed the increase of N_{eff}/m with a similar threshold field strength. This article guides a possible direction for feather investigation of SWCNT film's behavior in the high THz fields. The feather investigation of the samples with different diameters is required.

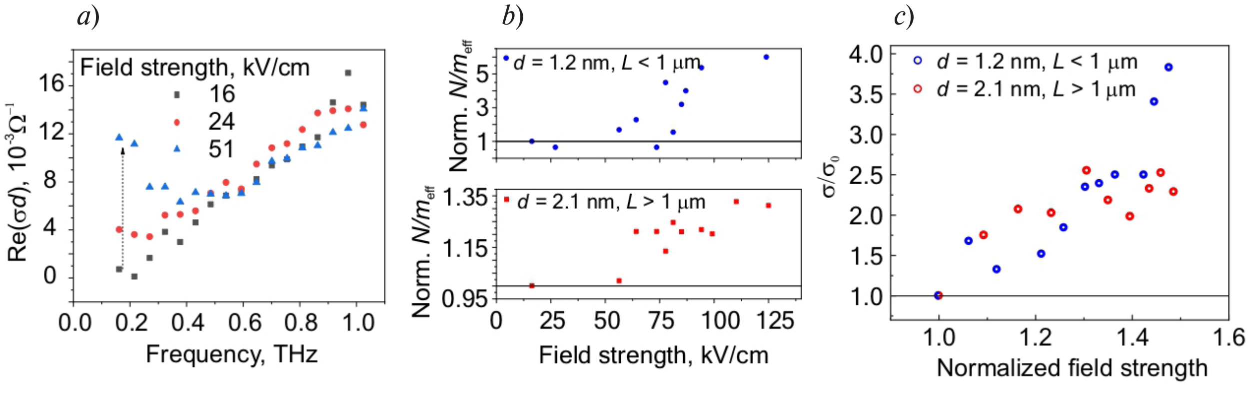

Fig. 1. Sheet conductivity of SWCNTs at different field strength (*a*); ratio of effective density of electrons to effective mass, N_{eff}/m, versus field strength (*b*); change in the conductivity ratio σ/σ_0 as a function of field strength (*c*)

Conclusion

In conclusion, strong THz pulses give rise to highly nonlinear conductivity in SWNTs. In particular, intense THz fields induce a large nonlinear dependence of N_{eff}/m obtained by using a model-independent approach. This theoretical analysis suggests that strong THz fields enhance the conductivity of the SWNTs, inducing strong nonlinear electron dynamics as the result of several competing processes influencing N_{eff}/m. However, we can't independently treat whether N_{eff} or m influenced such behavior. The results of this work can be used to predict the performance of CNT devices (modulators, polarizes, lenses, and so on) in the THz high-field.

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