Carrier dynamics in graphene studied by ultra-broadband optical-pump/terahertz probe spectroscpy

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***Abstract*—Carrier dynamics in single-layered graphene has been studied by reflection-type of THz time domain spectroscopic ellipsometry (THz-TDSE) and optical-pump/THz-probe spectroscopy (OPTP). We successfully determined the frequency-dependent complex sheet conductivity of photo-excited monolayer Graphene from 0.3 to 16 THz. By analyzing with Drude model, the time dependence of Drude weight and carrier scattering rate in Graphene were estimated. It is found that Drude weight decreases and carrier scattering rate increases after photo-excitation, which contribute to the negative photoconductivity in graphene.**

I. INTRODUCTION

Single-layered graphene is a gapless two-dimensional material with linear electron and hole bands. The charge carriers in this system behave like massless Dirac fermions and have high Fermi velocity and mobility. Because of such unique properties, graphene has many possibilities of application for electronic or optoelectronic devices. In order to realize such high performance devices, the understanding of the carrier dynamics of graphene is necessary. Although the ultrafast optical response of photo-excited graphene has been intensively studied, the carrier dynamics has not been fully understood yet.

In this study, we developed an ultra-broadband optical-pump/THz-probe (OPTP) spectroscopy by the combination of the air plasma and THz air breakdown coherent detection (THz-ABCD) to understand the carrier dynamics in graphene The system has ultra-broadband measurement capability from 0.5 to 30 THz and ultra-fast time resolution of 150 fs, which enables to perform the direct measurement of the time dependence of the optical complex conductivity associated with the intra-band transition in a single-layered graphene by using our OPTP spectroscopy system.

II. RESULTS

Figure 1(a) shows the temporal profiles of the photo induced reflectance changes with different optical fluences. The second peak appearing 2 ps after the photo excitation is due to the multiple reflection of the excitation pulses inside the PET substrate. The THz reflection decreases rapidly after photoexcitation, and then recovers to the original value with a relaxation time τ=1.4 ps obtained from the fitting assuming the double exponential decay for the double photoexcitation. Figure 1(b) and (c) show the complex optical conductivity $\sigma(\omega)$ of the unexcited graphene and the photo-excited one at 0.5 ps after the photo-excitation. The $\sigma(\omega)$ decreases rapidly by the photo-excitation, which quite differs from the response in conventional semiconductors. The frequency dependence of $\sigma(\omega)$ of the unexcited and photo-excited graphene clearly show the behaviour of the Drude model which is expressed by the eq. (1).

$$\sigma(\omega)=\frac{D}{\pi}\frac{1}{(\Gamma-i\omega)} \qquad (1),$$

here Γ is the average scattering rate for momentum changing collisions of charge carriers, D is Drude weight. For the degenerated electron gas condition in graphene, D is expressed by $D= N_c\ \pi e^3 v_F^2$ at zero Kelvin. Here, N_c is the carrier concentration and v_F is Fermi velocity of graphene. Fig. 2. shows the time dependence of D and Γ which were obtained from the fitting of $\sigma(\omega)$ with Drude model. After the photo excitation, D (proportional to N_c) decreases and Γ increases, which results in the negative photoconductivity of graphene. The increase of Γ is attributed to the increased optical phonon scattering due to the heating of carriers by photo-excitation [2]. In Ref. [2], the slight increase of D has been reported, which is explained by the heating of the carrier temperature after the relaxation of the photo-excited carriers via intra- and inter-band transitions to a state with a single chemical potential. However, the decrease of D induced by the photo-excitation cannot be explained by this story. This result suggests that further study is needed to understand this unusual carrier dynamics of graphene.

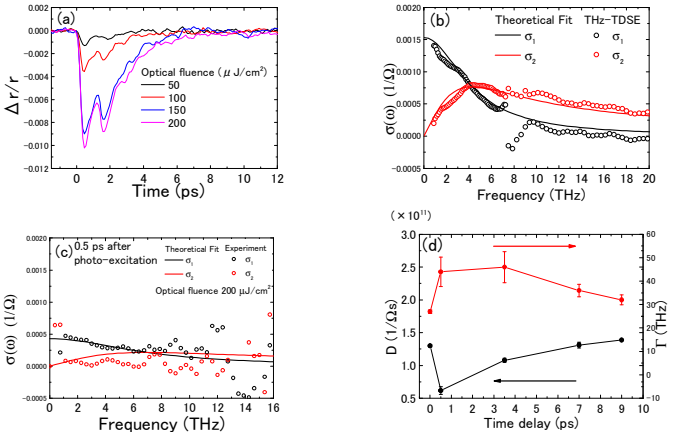

Fig. 1 (a) The time resolved reflectance change of photo-excited graphene. The $\sigma(\omega)$ of the single layered graphene (b) without the photo-excitation and (c) at 0.5 ps after the phto-excitation. (d)The temporal evolutions of the D and Γ after the photo-excitation in graphene.

REFERENCES

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