Ultrafast Detection of Carrier Relaxation in Graphene Quantum dots

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Abstract

Graphene quantum dots (GQDs) have been recently fabricated with the size smaller than 4 nm using acidic exfoliation and etching of pitch carbon fibers, leading to bandgap opening at the K-point [1]. While fabricated GQDs, however, one cannot expect pure GQDs due to oxidation or other functional groups. Therefore the carrier dynamics in GQDs is affected by those defects. For further optical and optoelectronic applications, it is important to investigate the dynamics of both photoexcited carriers and defects that might interrupt the carrier relaxation in GQDs. Transient absorption (TA) spectroscopic technique is one of the powerful tools to study the dynamics of carriers and defects [2,3]. Using femtosecond TA technique, we have studied the photoexcited carrier distribution, and the unusual photo-bleaching behavior by detects, such as hydroxyl groups, which occurred on the femtosecond time scale.

We performed TA spectroscopy with different excitation energies, tuned by using an optical parametric amplifier (OPA) which pumped by a Ti:sapphire regenerative amplifier of 1-kHz repetition rate. The pulse duration of OPA was around 200 fs, and 5 μ J energy per pulse were used. TA signal was probed by white light continuum (WLC) in visible range from 450 to 750 nm (1.65 eV~2.75 eV). In order to generate WLC, the Ti:sapphire amplifier with 800 nm (1.55 eV) center wavelength was focused on a sapphire window.

Figure 1 shows spatially and temporally resolved TA map in GQDs, excited above resonance and at resonance, respectively. Above resonance shown in Fig. 1(a), snapshots of hot carrier distribution in GQDs after laser excitation can be obtained by photo-induced absorption. At resonance shown in Fig. 1(b), the strong photobleaching behavior is shown in earlier delay time. The bleached energy state appears below 400 meV from the excitation energy. The energy difference corresponds to the energy of hydroxyl groups. It implies that the photoexcited carriers might be captured at the site of hydroxyl groups, and it takes less than 1 picosecond. In order to understand this behavior, we compared our data obtained from GQDs with that from large size of pristine graphene. We also annealed GQD sample at 200 $^{\circ}$ C, and 600 $^{\circ}$ C to remove the defects. Furthermore, we measured the excitation power dependence, and the carriers decay within 1 ~ 10 picosecond when the excitation power is higher

References

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Figures

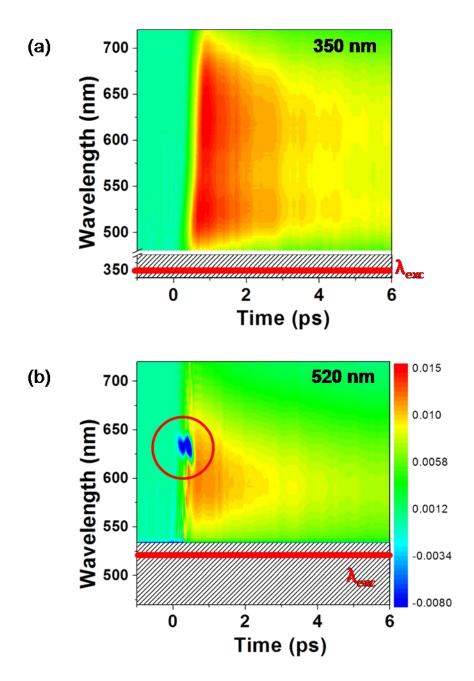


Figure 1 Spatially and temporally resolved transient absorption map in graphene quantum dots, excited (a) above resonance of 350 nm (3.54 eV), and (b) at resonance of 520 nm (2.38 eV), respectivley. 5 μJ energy per pulse were used.