

Femtosecond mid-infrared luminescence with hot-phonon effects in graphenes and graphite

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1. Introduction

Recently graphene has been attracting much attention in applications to high speed electronics such as field effect transistors, single electron transistors and optical sensors, because it has a unique symmetric linear dispersion called Dirac-cone for electron and hole, and high carrier mobilities. For these applications, interaction of the electrons with phonons is important, because the optical response is limited by cooling of the carriers. Transport property is also dependent on the speed of cooling of the hot carriers. From this view point, the dynamics of high energy electrons and coupled phonons have been investigated with various experimental methods, such as transient absorption [1], reflectance[2], anti-Stokes Raman scattering [3], photoelectrons[4,5], and luminescence[6,7]. In most of the reports, the response of photo-excited electrons (holes) has a fast (order of 100 fs) and a slow component ranging from 1.5 to 3 ps. The time constants strongly depends on the observation photon energy, excitation fluence, thickness (layer number), and sample condition. Large difference in the time constant has been reported for free standing and substrate-supported graphenes.

In most of the transient absorption measurements, the responses are observed at single energy in relatively high energy region (typically from 1 to 1.6 eV). To understand the whole picture of photo-excited electron dynamics, it is important to observe the response in a wide energy range especially at low energies close to Dirac point. Wider range measurements have been performed by luminescence between 1.5 and 3.5 [6] or between 0.7 and 1.4 eV [7]. Time and angle resolved photoemission spectra from -0.5 to 1.5 eV (measured from Fermi energy) including Dirac point has been reported recently [5]. However, in the last case, thickness dependence have not been reported. In spite of these efforts, the reported lifetimes are not consistent each other and especially the thickness dependence is controversial. In this report, we observed luminescence down to 0.3 eV, corresponding to 0.15 eV electron, with a time resolution of 170 fs on mono-, bi-, 6-8 layer graphenes and graphite which can be assumed as infinite layer graphene.

2. Experiment

We used mono-, bi- and 6-8-layer graphene sheets purchased from ACS MATERIAL® and HOPG graphite. The graphene sheets are transferred on to fused silica substrates.

The sample was excited at 1.57 eV (790 nm), by 70 fs pulses at a repetition rate of 200 kHz, and the infrared luminescence signal was up-converted to visible light and analyzed by a double grating monochromator and detected with a photon counting system.

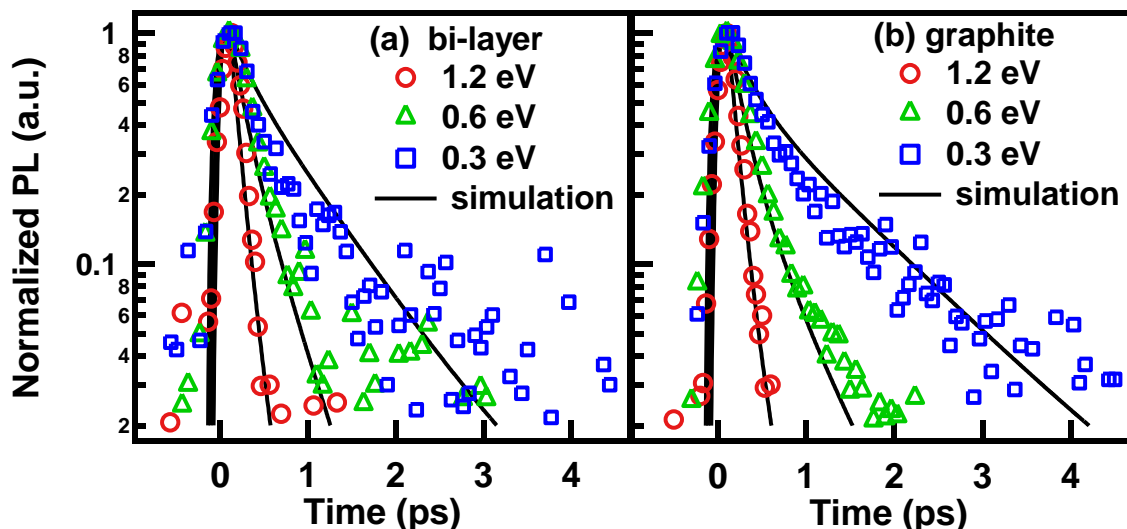


Fig.1 Time evolution of the luminescence intensity in bi-layer graphene (a) and graphite (b) observed at 0.3, 0.6, and 1.2 eV.

3. Results and Discussion

Photon energy dependences of the decay profiles in bi-layer graphene and graphite under relatively high excitation fluence (100 mW) are shown in Fig. 1(a) and (b), respectively. In graphite, the lifetime becomes longer at lower photon energy as reported in ref. [8]. In the case of bi-layer graphene, however, the lifetime at 0.3 eV is not so long as that in graphite, while the lifetime at 1.2 eV is almost the same. Elongation of the lifetime at lower energy is ascribed to slower decay of electron population at lower energy, as predicted by Fermi-Dirac distribution assuming cooling down of the electron system via optic phonons.

Luminescence signals from graphenes with different thickness are shown in Fig. 2. At the lowest energy 0.3 eV, the thickness dependence is most clearly seen, that is, the lifetime in mono-layer graphene is roughly one half of that in graphite, while that of 6-8 sample is almost the same as that of graphite.

We try to understand these observation in terms of the two-temperature model [7]. We assume that the energy of the electron system is transferred to the optic phonon (G mode, $\varepsilon_{op} = 0.2$ eV) as the first step and that the energy of the hot optic phonon is dissipated to the large heat bath within the graphene sheet. We can derive the differential equations for the energy of electron U_e and optical phonon U_{OP} .

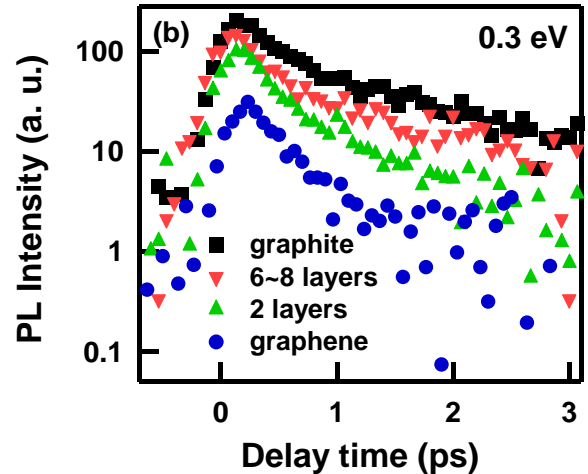


Fig. 2 Thickness dependence of the luminescence decay profiles observed at 0.3 eV. Data for 1, 2 and 6-8 layers graphene and for graphite are shown.

,where $G(t)$ is the envelop of pulse shape T_e , T_{OP} , and T_{RT} are electron temperature, optical phonon temperature, and temperature of the heat bath ($T_{RT} = 300$ K), C_{OP} is the heat capacity of the optical phonon, D_e is the density of state of electron, and f_e is the Dirac-Fermi function at T_e .

The calculated curves for graphite are shown in Fig. 1(b) by solid lines, which are in good agreement with experiment. Then the fast decays in bi-layer graphene are reproduced simply by adding a path from the optic phonons of the graphene or graphite to the optic phonons of the substrate, having an infinite heat capacity as shown in Fig. 1(a). This suggests that the interaction between the hot carriers and the substrate is the main cooling mechanism in silica-supported graphene as proposed in ref. [3].

References

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