Ultrafast Optical-to-electrical Conversion Processes in Graphene

Katsumasa Yoshioka, Taro Wakamura, and Norio Kumada

Abstract

Graphene photodetectors have gained significant interest due to their predicted high sensitivity and rapid electrical response to broadband light, offering the potential for superior performance compared with conventional semiconductor devices. However, the experimental evaluation of the operation speed under a zero-bias voltage has been hindered by challenges associated with device architecture and measurement instrumentation, restricting the characterization of graphene's intrinsic dynamics and clarification of its operating mechanism. This article presents an ultrafast graphene photodetector developed at NTT laboratories, detailing the measurement technology and operating mechanism.

Keywords: graphene, photodetector, terahertz electronics

1. Graphene as an optical-to-electrical conversion device

Photodetectors (PDs) play a crucial role in optical information communications and sensors by converting optical signals into electrical signals. Developing a PD that operates at high speeds for broadband optical signals creates new opportunities for advanced information processing through optical sensors and receivers working across a wide range of frequencies. This requirement can be satisfied with graphene, a promising material that exhibits high efficiency with its ability to absorb 2.3% of incident light with a single atomic layer and operate in an ultrawide frequency band, from terahertz to ultraviolet light [1]. The photothermoelectric (PTE) effect enables graphene PDs to operate at zero bias, which is essential for low power consumption and improved signal-tonoise ratio. However, previous research on graphene PDs has limited the experimental operating speed under zero bias to around 70 GHz [2], far from the theoretical expectation of 200 GHz. The limitations in device structure and measurement systems have hindered the investigation of the intrinsic response of graphene and the clarification of its operating mechanism, leading to a lack of design guidelines for maximizing the performance of graphene PDs. Therefore, the demonstration of 200-GHz operation speed and the elucidation of the physical properties of graphene, including the optical-to-electrical (OE) conversion mechanism, is crucial for further developments in graphene PDs.

2. Ultrafast photocurrent detection based on a novel device architecture and measurement approach

Ultrafast OE conversion in a graphene PD requires a device design that facilitates prompt tracking of incident light signals and a measurement technique capable of high-speed photocurrent detection. To meet these requirements, we used a zinc oxide thin film as a gate material to eliminate the current delay caused by capacitive coupling (**Fig. 1(a**)) and used on-chip terahertz spectroscopy [3] for photocurrent readout (**Fig. 1(b**)). The OE conversion efficiency of graphene PDs using the PTE effect varies significantly with the charge density. Thus, a gate structure must be integrated into the device. Because the gate electrode must possess high conductivity, gold is commonly used as the gate material; however, this limits the operating speed to below 100 GHz due to



(a) Schematic of a graphene PD. Graphene is sandwiched between hexagonal boron nitride (hBN) as an insulator, and connected to metal source and drain electrodes. After being covered with aluminum oxide (Al₂O₃) insulating film on the surface, zinc oxide (ZnO) is deposited on top as a gate electrode. (b) Microscope photograph of an on-chip terahertz-spectroscopy circuit. When pump light is applied to the graphene PD, the resulting photocurrent is propagated through the drain electrode and detected by illuminating the PC switch with probe light.

Fig. 1. Measurement system for ultrafast OE conversion.

unwanted capacitive coupling. We addressed this issue by using a zinc oxide thin film, which enables control of the high-frequency response by controlling growth conditions. This material possesses a unique combination of being a good conductor for directcurrent signals while being transparent at terahertz frequencies [4]. As a result, we successfully eliminated the capacitive coupling-induced current delay. The high-speed oscilloscopes commonly used to read out the photocurrent struggle to measure response faster than 100 GHz because of the bandwidth limitation of the electronics. However, we overcame this challenge by using on-chip terahertz spectroscopy, which enabled us to measure the ultrafast response of the graphene PD with a measurement bandwidth approaching 1 THz through the detection of photocurrent using an on-chip photoconductive (PC) switch. The photocurrent generated by irradiating the graphene PD with a pump pulse (femtosecond laser pulse) propagates through the drain electrode to the PC switch (Fig. 1(b)). Because the PC switch becomes conductive when irradiated by the probe pulse (also femtosecond laser pulse), the signal flows to the ammeter only when the generated photocurrent and probe pulse overlap in time. Therefore, measuring the photocurrent waveform with extremely high time resolution is possible by constantly changing the time difference between the pump and probe pulse. Our innovative device architecture and measurement approach have enabled us to explore the ultrafast OE conversion mechanisms in graphene, which was previously unachievable using conventional methods.

3. Overview of the OE conversion processes in graphene

The photocurrent waveform of the fastest response among measured signals is shown in **Fig. 2(a)**. The various frequency components of the photocurrent obtained by the Fourier transform (**Fig. 2(b**)) show that the 3-dB bandwidth of the graphene PD reaches 220 GHz, more than three times the 70 GHz reported previously and exceeding the theoretical expectation of 200 GHz. This is the first successful extraction of the intrinsic response of graphene unimpeded by device structure or measurement method. To investigate the physical process that determines the dynamical response of the photocurrent, we first focused on the decay time of the photocurrent by fabricating multiple devices with different graphene mobilities.



(a) Time evolution of photocurrent due to optical pulse illumination. (b) The magnitude of each frequency component of the photocurrent obtained by the Fourier transform. The operating speed is 220 GHz, where the current magnitude drops by 3 dB.

Fig. 2. Verification of operating speed of 220 GHz.



(a) Seebeck coefficient (an indicator of sensitivity). Sensitivity becomes higher when the electron scattering in graphene is small and mobility is high. (b) Decay time (inverse of operating speed). As mobility increases, the scattering decreases and decay time increases (speed decreases).

Fig. 3. Tradeoff between detection sensitivity and operating speed.

Figure 3 shows that lower graphene mobility is associated with shorter decay times. This result suggests that the decay of the photocurrent in graphene is caused by a decrease in the electron temperature in

graphene, which is increased by light irradiation. Because of collisions between acoustic phonons and defects, a sample with lower mobility and more defects loses more energy in a single collision event,



(a) As the carrier density in graphene decreases, the time the current peaks after light irradiation becomes 4 ps later.
(b) A schematic of the measurement configuration. The current is extracted from the lower drain electrode.
(c) The peak shift does not depend on the length or spot position of the graphene.

Fig. 4. Dependence of photocurrent-response time on charge density.

resulting in faster decay times. However, the Seebeck coefficient, which corresponds to PD sensitivity, increases as mobility increases (Fig. 3), so there is a tradeoff between bandwidth and sensitivity in graphene PDs. Therefore, it is necessary to select optimal graphene mobility in accordance with the intended application.

To identify the mechanism that determines the photocurrent generation and propagation response (**Fig. 4(a)**), we investigated the dependence of the photocurrent on the charge density. The results indicate that a decrease in charge density produced a delay in the peak position of about 4 ps. Such a significant peak shift is unprecedented and provides important clues for understanding the OE conversion processes. We can consider two possible causes: 1) the propagation time of the photocurrent through graphene changes or 2) the time for photocurrent generation after light irradiation changes. To test the first possibility, we measured the peak shift for various device lengths and the irradiation position of the pump light (Fig. 4(b)). All the results fall on a single curve (Fig. 4(c)), indicating that the photocurrent propagation time in graphene is too short to be measured, so the cause of the peak shift must come from the change in the photocurrent-generation time. The ultrashort propagation time in a graphene PD results from its gapless nature. The absence of a bandgap in graphene allows the electric field created by light irradiation to propagate at the speed of light, instantly displacing the electrons near the electrode and generating an immediate flow of current without the need for propagation. The time variation in photocurrent generation is due to the PTE effect, which transforms the electron temperature into a voltage. This means that the non-equilibrium electron state created by the light irradiation quickly settles into a Fermi-Dirac distribution through intraband electron scattering. Our results also indicate that the time it takes for thermal equilibrium is highly dependent on the charge density of the graphene. These findings provide a comprehensive understanding of the ultrafast OE conversion processes in graphene, including the generation, propagation, and decay of photocurrent upon light irradiation [5].

4. Future development

The results presented in this article indicate the high potential of graphene as a material for broadband, high-speed PDs. However, our fabrication process of manually exfoliating graphene from graphite is not suitable for mass production. While the quality of graphene deposited over a large area has been historically inferior to that obtained through exfoliation, the gap is closing as deposition techniques continue to improve. Future studies will focus on evaluating PDs that use large-area graphene to facilitate mass production. Additionally, research in the field of creating novel materials by stacking graphene and other two-dimensional materials could be applied to further enhance the operating speed of PDs. The results of this study could be used to gener-



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He received a B.E., M. Sc., and Ph.D. in science from the University of Tokyo in 2010, 2012, and 2015. He joined NTT Basic Research Laboratories in 2019 and constructed a semi-automatic heterostructure fabrication system with novel two-dimensional materials including graphene and transition-metal dichalcogenides (TMDs). His current interests are broad, and he has recently been working on nonreciprocal transport in noncentrosymmetric TMDs. In addition to his own projects, he also contributes to collaborative works for developing novel PDs based on twodimensional materials such as graphene. He is a member of the Physical Society of Japan. ate, control the propagation of, and detect graphene plasmons in the terahertz range. This opens up the possibility for on-chip handling of plasmon signals, bringing us closer to the goal of creating plasmonic circuits.

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