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Propagation properties of longitudinal wave in defect-free multilayer graphene at low temperatures and high magnetic field

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1. Introduction

Graphene has attracted great interest for long years because of its unusual electronic, magnetic, and mechanical properties[1]. Two-dimensional materials have been discovered, characterized, and applied since the discovery of graphene's extraordinary physical properties[2]. Many studies have been reported on the mechanical properties of multilayer graphene, including its interlayer interactions[3,4]. This interlayer interaction is considered to rely on the van der Waals force, and this relatively weak coupling force presents the possibility of an entirely new device. However, its origin, theoretical modeling, and calculation are still under investigation. To discuss them, elastic constants are very important because it directly reflects the interaction between atoms and layers — the van der Waals force —. Recent progress to synthesize defect-free multilayer graphene[5,6] enables us to accurately measure the out-of-plane elastic constant [7].

Moreover, graphene exhibits a unique property under a magnetic field. When a magnetic field is applied along the *c*-axis, the cyclotron motion of the free electrons in graphene is excited and causes quantized energy levels, that is the Landau level. The electronic state is modified by the applied magnetic field, and the van der Waals force is also predicted to depend on the electronic state because it stems from instantaneous dipole and exchange interactions. However, The magnetic field dependence of mechanical properties such as interlayer interactions and phonon-phonon interactions has not been demonstrated to our knowledge.

In this study, we measure the propagation dynamics of high-frequency phonons in defect-free multilayer graphene using picosecond ultrasonics (PU). We develop optics for performing the PU measurement at low temperatures under a high-magnetic field using a superconducting-magnet chamber, which allows us to investigate the interaction between phonon-propagation and electron state with Landau levels.

2. Experiments

The defect-free multilayer graphene was synthesized by beating polyimide film under in-plane tension[6], and its thickness is ~2.1 µm. As shown in the inset of **Fig. 1**, we keep the multilayer graphene free-standing by transferring it to a Si substrate with many holes. The temperature control

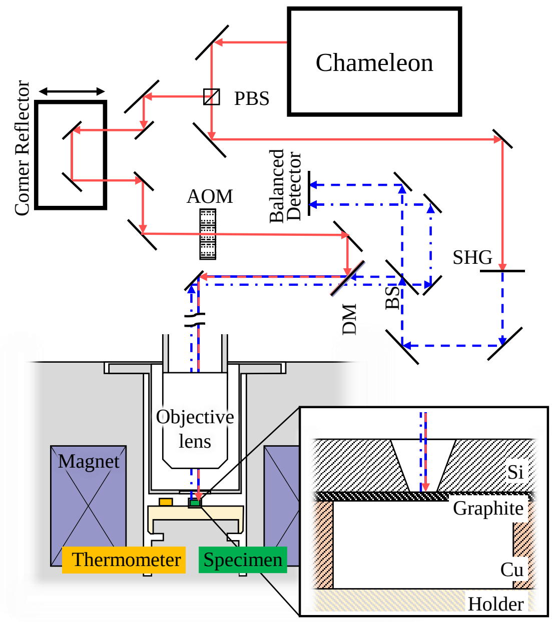

Fig. 1 Schematic of the optical systems. Red and Blue lines denote pump and probe light, respectively. The inset shows the Graphene installation and a cross-sectional view of the surrounding area.

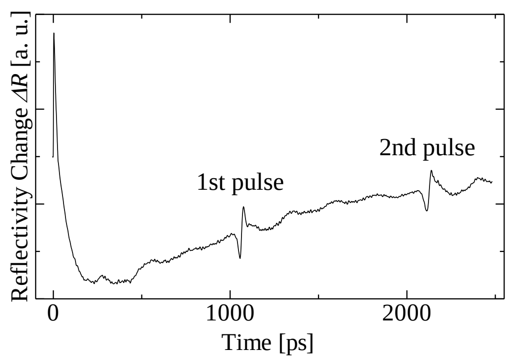

Fig. 2 Reflectivity changes at 28 K and 0 T. The value t = 0 corresponds to the time when the strain pulse is excited. Define the first and second pulses as shown in the figure.

of the multilayer graphene specimen was achieved by attaching a Cu plate.

We show the schematic of the optics in Fig. 1. We use a titanium-sapphire pulsed laser, whose pulse width, repetition rate, and center wavelength are under 140 fs, 80 MHz, and 800 nm, respectively.

Light modulated at 100 kHz by the acousto-optic crystal modulator (AOM) is focused onto the specimen surface to excite electronic levels and cause thermal expansion, leading to a strain pulse. Another light at 400 nm wavelength generated by a second harmonic generator (SHG) is focused on the specimen surface to detect transient reflectivity changes. At this time, the two types of pulse light enter with time differences controlled by the optical delay path.

The specimen was placed in the center of a superconducting magnet at 28 K. The strain pulse, generated by the thermal expansion with electronic excitation is distorted due to nonlinearities and decays due mainly to phonon-phonon interactions while propagating in the thickness direction. An example of the observed reflectivity change is shown in **Fig. 2**. 0 ps corresponds to the time when the reflectivity reached its maximum value. The arrival time between the first and second pulses is used to evaluate the interlayer interaction.

Frequency analysis was made by extracting data around each pulse and applying the fast Fourier transform (FFT). The obtained FFT spectra are shown in **Fig. 3**. By comparing the amplitudes of first and second echoes at each frequency, we can evaluate the attenuation coefficient.

Furthermore, the superconducting magnet generates the Landau level by applying a magnetic field of 0–5 T. Thus, we investigate the dependence of interlayer interactions and phonon-phonon interactions on Landau levels.

3. Discussion

The wavelength of the pump light is 800 nm, and its penetration length is about 50 nm, which is about 2.9% of film thickness. As a result, the analytical data is expected to show interlayer and phonon-phonon interactions with electrons filled to the Fermi level while the Landau levels appear. Therefore, the magnetic field dependence of the elastic constants can be attributed to changes in the coupling force and magnetization direction due to the occurrence of Landau levels.

In addition, the scattering attenuation of ultrasound caused by defects could be negligible since our sample includes very few defects. Therefore, we can evaluate the anharmonicity which causes the phonon-phonon interaction attenuation by analyzing the attenuation of acoustic pulses.

From the experiments, we find a tendency for

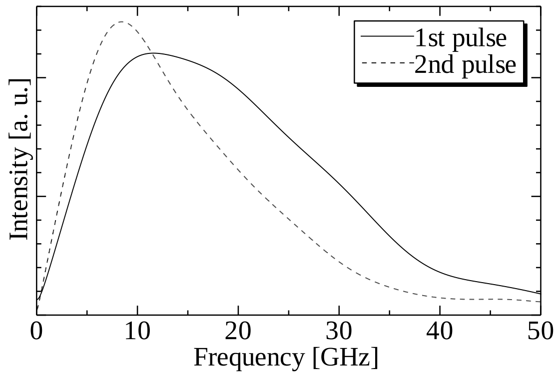

Fig. 3 Corresponding FFT spectra. Solid and dashed lines represent the spectra of 1st and 2nd pulses, respectively.

the elastic constant to decrease as the magnetic-field strength increases. Furthermore, we found that phonons in the high-frequency band of several tens of GHz attenuated comparatively a lot with propagation. We will further investigate their effects precisely.

4. Summary

In conclusion, we have demonstrated the propagation of strain pulses in Landau levels. The Landau level can be varied by changing the intensity of the magnetic field. This shows the dependence of the mechanical properties on the electronic state. This could lead to further clarification of van der Waals forces.

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