論文の内容の要旨

Ultrafast nonlinear optical responses of Landau-quantized graphene in the terahertz range
（ランダウ量子化した単層グラフェンのテラヘルツ帯超高速非線形光学応答）

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Electrons in graphene, a single atomic layer of carbon atoms forming a hexagonal lattice structure, exhibit the linear energy dispersion in the low-energy regime and mimic massless relativistic particles, that is, massless Dirac fermions. This relativistic property makes graphene unique among condensed-matter materials. When a strong magnetic field is applied perpendicularly to graphene, unlike the equidistant Landau levels (LLs) in ordinary two-dimensional electron gas (2DEG), the energy spectrum is separated into LLs with non-equidistant energy spacing covering the THz and mid-infrared range, which is given by $\varepsilon_n = \mathrm{sgn}(n) v_F \sqrt{2|e|\hbar B|n|}$ (n = 0, ±1, ±2…), where v_F is the Fermi velocity, e is the elementary charge, B is the magnetic field and n is the LL index. The unique electronic properties have motivated intensive studies on the magneto-optical responses of Landau-quantized graphene. Intriguing linear magneto-optical responses specific to the massless Dirac electrons have been observed in the THz and the mid-infrared range such as the unusual optical transition selection rule between the LLs and quantum Faraday and Kerr rotations in the quantum Hall regime.

In 2DEG systems under strong magnetic fields, the cyclotron orbits are confined within the two-dimensional plane with the radii on the order of the magnetic length l_B. Therefore, the optical transition dipole moments between LLs are characterized by el_B and become so large. Then, the extreme nonlinear optics regime, where the Rabi frequency, $\Omega_R = \mu E/\hbar$ (μ is the transition dipole moment and E is the electric field strength), exceeds the carrier frequency of the light pulse, may be realized with feasible electric field strength in the THz range. In such a strong light-matter coupling regime in atomic or molecular systems, the perturbation theory breaks down and a number of fascinating phenomena emerges such as carrier-wave Rabi flopping, high-order harmonic generation (HHG), and attosecond pulse generation. Recent advances of carrier envelope phase-locked intense THz and mid-infrared light sources have allowed us to get access to such a strong coupling regime even in solid states, as exemplified by sub-cycle coherent control of electrons and HHG in semiconductors. In these experiments, ultra-intense electric fields of ~10 MV/cm were needed to explore such nonlinear optical phenomena. From this point of view, a Landau-quantized 2DEG system may offer a unique condensed-matter playground to study the non-perturbative light-matter interaction phenomena at much weaker electric fields.

However, in ordinary 2DEG systems with equidistant LLs, the optical nonlinearity cannot show up unless anharmonicity is induced into the system such as the non-parabolic electron band dispersion and electron-ion interaction effect. In contrast, the LLs in graphene intrinsically exhibit anharmonicity originating from the non-equidistant energy spectrum, reflecting the relativistic nature of electrons. Indeed, the coherent nonlinear optical responses of the non-equidistant LLs in graphene have attracted many theoretical interests. However, only a few time-resolved spectroscopies have been performed concerning the non-equilibrium dynamics of the LLs in graphene such as the carrier relaxation, and the coherent nonlinear optical responses in the Landau-quantized regime remain unexplored.

In this dissertation, we aim to reveal the nonlinear optical responses of Landau-quantized graphene in the THz range in the strong light-matter coupling regime. We first perform single pulse transmission experiments using intense THz pulses. We find that the magnitudes of the Faraday rotation angle and ellipticity spectra are suppressed with increasing the peak field of the incident THz pulses. The Faraday rotation almost vanishes in the case of the incident THz pulse with the peak field of E_{peak}=25 kV/cm. To further investigate the observed nonlinear THz responses, we develop a THz pump-THz probe magneto-optical spectroscopy system under the strong magnetic fields. The pump-probe experiments are performed on a monolayer graphene epitaxially grown on the Si face of 4H-SiC at the applied magnetic field of B = 3 T, where the relevant LL transition energy between the LL for n = 5 (LL_5) and LL_6, ΔE_{65}=13.7 meV, is ~3 times larger than the central photon energy (5 meV) of the pump THz pulse with the peak field of 45 kV/cm, as schematically shown in Fig. 1. Despite the far off-resonant excitation conditions, we observe the ultrafast suppression and recovery of the magnitudes of the Faraday rotation angle $\theta_F(\omega)$ and ellipticity $\eta_F(\omega)$ spectra under the irradiation of the pump THz pulse shown in Fig. 2 (a). By extracting the pump-induced change of the conductivity spectra $\Delta\sigma_\pm(\omega)$ for the right- (+) and left- (-) handed circularly polarized components of the probe THz pulse, we elucidate that the nonlinear THz Faraday rotation results from an absorption bleaching and induced absorption for the right- and left-handed circularly polarized component, respectively, as shown in Fig. 2 (b). This finding indicates that the far off-resonant intense pump THz pulse strongly drives the carrier distribution and a drastic LL-ladder climbing of electrons occurs not only in the conduction band but also even in the valence band far below the Fermi energy.

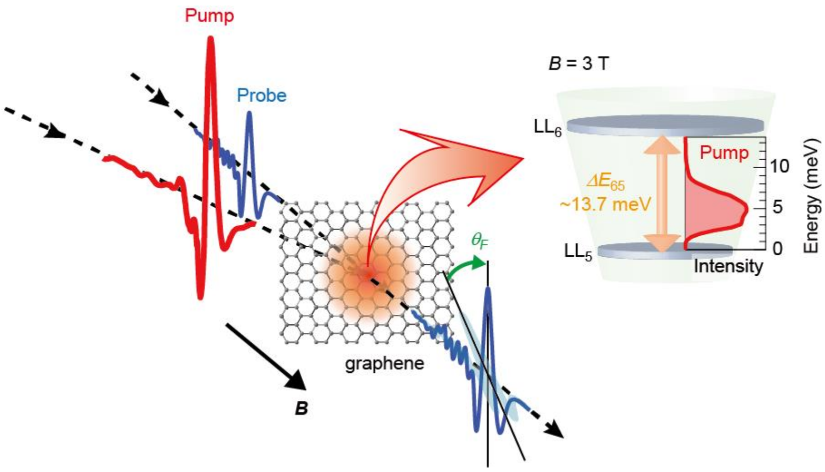

Figure 1: Schematic of the experimental configuration in the THz pump-THz probe experiments on Landau-quantized graphene. Pump THz spectrum is shown in comparison with ΔE_{65} at B = 3 T. θ_F denotes the Faraday rotation angle.

In order to study the microscopic origin and mechanism of the observed THz nonlinearity under the irradiation of the pump THz pulse, we perform numerical simulations based on the density matrix formalism without using rotating-wave approximation. The pump-induced drastic population redistribution expected from the experimental results is simulated by the numerical calculations (Fig. 2 (c)), where the maximum Rabi frequency associated with the LL_5-LL_6 transition is estimated to be $\hbar\Omega_R \sim 15$ meV, which is ~3 times larger than the central pump photon energy of 5 meV and even larger than the LL energy spacing of ΔE_{65}=13.7 meV. Accompanying the drastic population change, the macroscopic polarizations between the LLs are found to be strongly driven so that the polarizations between the LLs far from the Fermi energy also start to oscillate.

Finally, by using the numerical simulation, we investigate the experimental feasibility for detection of the higher harmonics generated from such nonlinear macroscopic polarizations between the non-equidistant LLs in graphene under the irradiation of multicycle THz pulses. The peak electric field of the multicycle THz pulse for the detectable higher harmonic generation is estimated to be several tens of kV/cm and expected to be feasible with recent intense THz generation techniques.

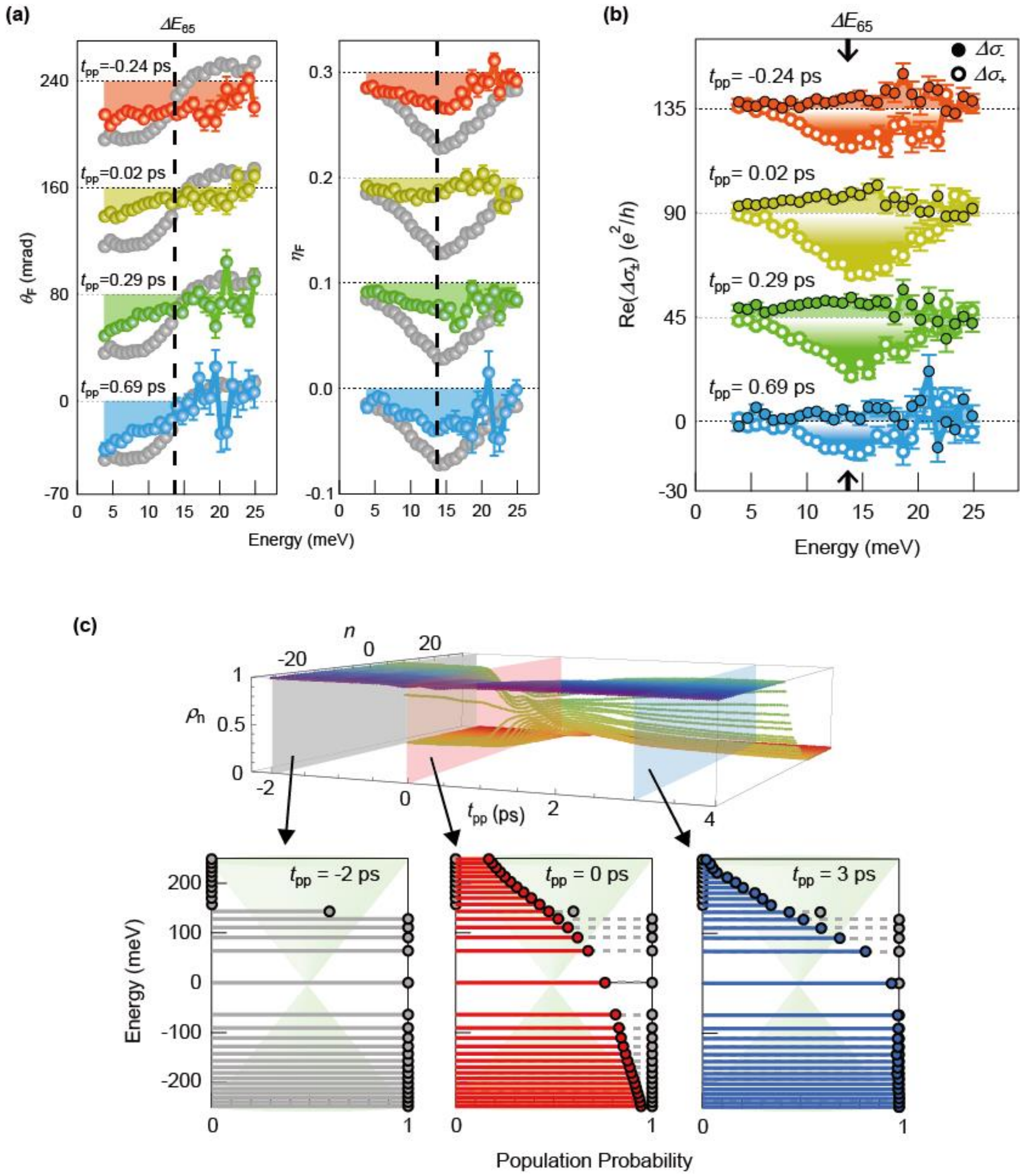

Figure 2: (a) Faraday rotation angle (left panel) and ellipticity (right panel) spectra at different pump-probe delay times t_{pp}. Gray circles represent the spectra without the pump excitation. The dashed lines show ΔE_{65}. (b) Real parts of the pump-induced change of the conductivity spectra for right- ($\Delta\sigma_{+}$) and left- ($\Delta\sigma_{-}$) handed circularly polarized light represented by open and solid circles, respectively, at different pump-probe delay times t_{pp}. The black arrows show ΔE_{65}. In both (a) and (b), the horizontal dotted lines at each pump-probe delay time correspond to zero and the error bars are the standard error of the averaging process. (c) Calculated population probability distributions (lower panel) at t_{pp} = -2, 0 and 3 ps as indicated by the slices of the 3D plot of the population dynamics of LLs at the corresponding t_{pp} (upper panel). The gray dashed lines depicted in the lower panel represent the population probability distribution at t_{pp} = -2 ps.