Controlling Dirac Carrier Dynamics in Graphene via Phonon Pumping

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Abstract: Using time- and angle-resolved photoemission spectroscopy, we find that resonant excitation of the in-plane E_{1u} lattice vibration in epitaxial bilayer graphene leads to a decrease of the relaxation time associated with electron - optical phonon coupling.
OCIS codes: 320.7130, 320.7150, 160.3900, 160.6000, 240.6490.

Phonon pumping is a powerful tool to control the electronic properties of different strongly correlated materials on an ultrafast time scale [1]. Here, we apply a similar approach to control the dynamics of Dirac carriers in epitaxial graphene samples. Despite the absence of strong correlations in graphene, the coupling of the Dirac carriers to optical phonons and to plasmons is strong enough to result in a significant renormalization of the linear electronic structure [2]. As graphene monolayers (MLs) do not possess infrared (IR) active lattice vibrations, we have excited graphene bilayers (BLs) at various wavelengths in the mid-infrared range (both on- and off-resonance with the IR active E_{1u} in-plane vibration at 6.3 μm) and observed the response of the electronic structure with time- and angle-resolved photoemission spectroscopy.

Figure 1a shows the photocurrent for negative pump-probe delays for BL graphene at the $\overline{\text{K}}$-point of the Brillouin zone. The pump-induced changes of the photocurrent at the peak of the pump-probe signal for $\lambda_{\text{pump}} = 6.3\,\mu$m are displayed in Fig. 1b. Due to a small p-type doping of our graphene samples, the excitation wavelength is too small to generate electron-hole pairs. Therefore, the excitation is dominated by free carrier absorption [3] and the pump-probe signal mainly reflects the generation of hot electrons (indicated by a broadening of the Fermi cut-off) accompanied by a broadening of the band structure.

For quantitative analysis the photocurrent has been integrated over the angle range between the two dashed lines in Fig. 1a (see Fig. 1c) and fitted with a Fermi-Dirac distribution in the vicinity of the Fermi level. The fit result for the Fermi width (in case of ideal energy resolution this would directly correspond to the electronic temperature T_e) is shown in Fig. 1d. A double exponential fit accounting for the coupling of electrons to optical phonons (τ_1) and the emission of acoustic phonons (τ_2) describes the time-dependence of the broadening of the Fermi cut-off reasonably well. Similar data sets have been obtained for different λ_{pump} between 4 and 9 μm. Within the error bars, $\tau_2 = 2.8 \pm 1.2$ ps is independent of λ_{pump}. Both the maximum T_e (Fig. 1 e) as well as the fast relaxation time τ_1 (Fig. 1 f), however, exhibit a clear minimum at 6.3 μm at resonance with the E_{1u} vibration.

All pump-probe experiments have been done at an excitation fluences of $F = 0.26$ mJ/cm^2. Assuming that the whole pump energy goes into the electronic system, the maximum T_e (before dissipating energy to other degrees of freedom) should be proportional to $F^{1/3}$ [3], i.e., for the same F one would expect the same maximum T_e independent of λ_{pump}. Hence, the observed minimum of T_e as a function of λ_{pump} might either arise from the fact that part of the excitation energy goes directly to the phonon bath, or that the energy dissipates from the electronic system on a time scale shorter than the pulse duration of 30 fs.

The origin of the observed drop of the fast relaxation time at resonance with the E_{1u} phonon cannot be explained by a classical two-temperature model where $\tau \propto T_e^{-3}$ [5]. On the other hand, pump-induced changes of the electronic dispersion or non-linear coupling to other phonon modes of the system may play an important role. Despite the well-known failure of the adiabatic Born-Oppenheimer approximation [6], frozen phonon calculations have been used to predict a band gap opening at the $\overline{\text{K}}$-point [4]. However, within the energy resolution of our experiment, the electronic dispersion seems to be unaffected by the pump.

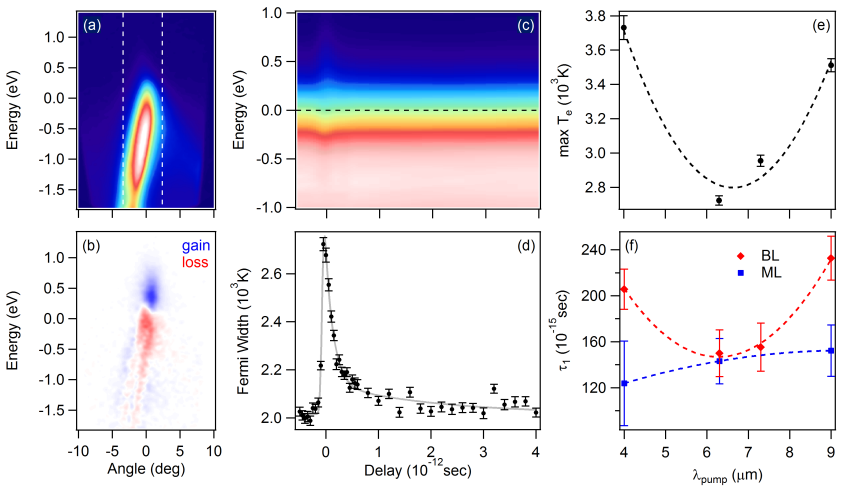

Fig. 1. (a) Photocurrent of bilayer graphene at negative time delays at the $\overline{\text{K}}$-point of the Brillouin zone. Dashed lines indicate the range of angle integration in panel c. (b) Pump-induced changes of the photocurrent at the maximum of the pump-probe signal for a pump wavelength λ_{pump} of 6.3 μm. (c) Angle-integrated photocurrent around the Fermi level as a function of pump-probe delay. (d) Width of the Fermi-Dirac distribution as a function of pump-probe delay (data points) together with a double exponential fit (continuous line). (e) Maximum electronic temperature at the peak of the pump-probe signal as a function of pump wavelength. (f) Dependence of the fast relaxation time τ_1 on pump wavelength in monolayer (blue squares) and bilayer graphene (red diamonds). The dashed lines in panels e and f are guides to the eye.

A comparison with ML graphene, where the in-plane mode cannot be directly excited with light, provides further clues about the mechanism behind the observed decrease of τ_1. We find that, in the ML, τ_1 is independent of λ_{pump} and always fast (blue squares in Fig. 1f). This might indicate that resonant excitation of the E_{1u} mode in BL graphene leads to a decoupling of the two graphene layers, possibly via non-linear phonon-phonon interaction between the E_{1u} mode and the shear mode of BL graphene [7]. Further optical pump-probe experiments are under way for a detailed investigation of non-linear phonon-phonon interactions in the BL system.

In summary, we have shown that via resonant excitation of the E_{1u} phonon in BL graphene we are able to control the relaxation dynamics of hot Dirac carriers, indicating that control via phonon pumping is also possible in systems without strong correlations and the associated competing ground states.

References

1. M. Rini *et al.*, "Control of the electronic phase of a manganite by mode-selective vibrational excitation," Nature **449**, 72–74 (2007); D. Fausti *et al.*, "Light-Induced Superconductivity in a Stripe-Ordered Cuprate," Science **331**, 189–191 (2011); A. D. Caviglia *et al.*, "Ultrafast Strain Engineering in Complex Oxide Heterostructures," Phys. Rev. Lett. **108**, 136801 (2012)
2. A. Bostwick *et al.*, "Quasiparticle dynamics in graphene," Nature Physics **3**, 36–40 (2007); A. Bostwick *et al.*, "Observation of Plasmarons in Quasi-Freestanding Doped Graphene," Science **328**, 999–1002 (2010)
3. I. Gierz *et al.*, "Snapshots of non-equilibrium Dirac carrier distributions in graphene," Nature Materials **12**, 1119–1124 (2013)
4. E. Cappelluti, and G. Profeta, "Hopping-resolved electron-phonon coupling in bilayer graphene," Phys. Rev. B **85**, 205436 (2012)
5. P. B. Allen, "Theory of Thermal Relaxation of Electrons in Metals," Phys. Rev. Lett. **59**, 1460 (1987)
6. S. Pisana *et al.*, "Breakdown of the adiabatic Born-Oppenheimer approximation in graphene," Nature Materials **6**, 198–201 (2007)
7. M. Först *et al.*, "Nonlinear phononics as an ultrafast route to lattice control," Nature Physics **7**, 854–856 (2011)