Frontiers of sub-cycle terahertz science: the fast, the strong and the small

M. Hohenleutner, F. Langer, O. Schubert, S. Baierl, T. Maag, C. Lange, M. Porer,
C. Pöllmann, P. Steinleitner, M. Eisele, M. A. Huber, M. Plankl, T. L. Cocker, and R. Huber
Department of Physics, University of Regensburg, 94030 Regensburg, Germany

***Abstract*—Phase-locked few-cycle pulses in the ultrabroadband terahertz (THz) spectral window[1-4] have become a powerful tool to access low-energy elementary dynamics in solids, on time scales shorter than a single oscillation cycle of light[5-14]. Here, we tackle the role of excitons and their dynamics in modern dichalcogenide monolayers and exciton-polariton condensates[7-9]. Furthermore, intense THz pulses are used as atomically strong bias fields to explore high-harmonic generation by dynamical Bloch oscillations in bulk solids[11-13]. Finally, we develop a novel microscope that introduces sub-cycle resolution on the few-nm length scale[14]. The experiments reviewed here challenge modern quantum theories and spark hope for electronics and magnetic storage at optical clock rates.**

I. Introduction

The possibility to generate precisely defined multi-octave-spanning waveforms throughout the entire THz, far- and mid-infrared spectral region has heralded a revolution in ultrafast optics. Latest THz sources[1-4], in combination with electro-optic detectors sampling the actual oscillating carrier wave of light at ever higher frequencies, have profoundly changed the way scientists explore light-matter interaction: It is now routinely possible to manipulate and monitor the quantum dynamics of the elementary constituents of condensed matter during a time span shorter than a single oscillation cycle of infrared lightwaves[1-14]. THz sub-cycle physics[5] has, therefore, developed into an extremely dynamic research field. While weak multi-THz transients can be used to probe ultrafast dynamics of a plethora of low-energy elementary excitations[2,3] ranging from phonons, via plasmons, magnons, and intra-excitonic transitions[7,8] to correlation induced energy gaps[9,10], the achievement of atomically strong THz fields[3,15] has opened new doors towards lightwave electronics. Finally, there has been intense effort to improve the spatial resolution of THz microscopy[14,16-18]. In this article, we review our recent work exploring three scientific frontiers that are concerned with (i) sub-cycle probing of novel low-energy dynamics in solids[7-9], (ii) THz-driven lightwave electronics in bulk semiconductors[11-13] and (iii) nanoscale near-field microscopy with sub-cycle time resolution[14].

II. Probing Sub-Cycle Dynamics in the Mid Infrared

In the first set of experiments, we exploit ultrabroadband phase-locked terahertz transients and high-sensitivity electro-optic detection[19,20] to probe atom-like internal quantum transitions of excitons – Coulomb bound electron-hole pairs – in transition metal dichalcogenides (TMDCs) and semiconductor microcavities. Monolayer TMDCs belong to the most

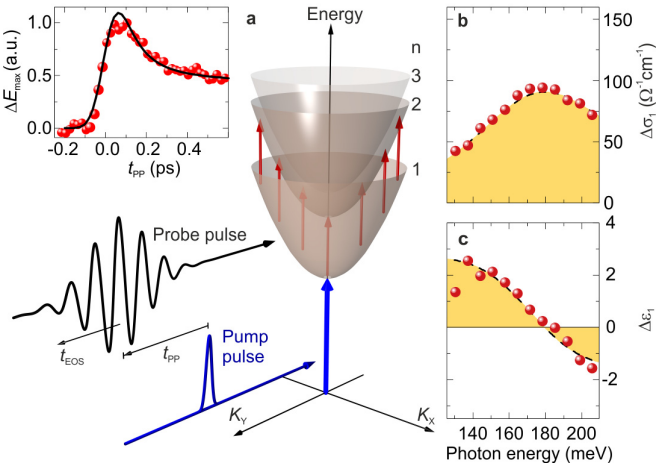

Fig. 1. Intra-excitonic spectroscopy of single-layer WSe_2. a, 1s excitons are resonantly generated by an optical pump (blue) while a multi-THz transient (black) delayed by t_{PP} probes the internal 1s-2p transition (red arrows) of all excitons, irrespective of momentum K_X and K_Y. t_{EOS}: electro-optic sampling time. Inset: ultrafast dynamics of exciton density for $\Phi = 41$ μJ/cm^2. Red spheres: Experimental data. Solid curve: Theoretical model accounting for ultrafast radiative decay (within 150 fs) and Auger recombination (few-ps scale). **b, c,** The real part of the pump-induced mid-infrared conductivity $\Delta\sigma_1(\hbar\omega)$ (**b**) and the corresponding real part of the dielectric function, $\Delta\varepsilon_1(\hbar\omega)$ (**c**) exhibit the characteristic shape of the 1s-2p exciton resonance (transition energy: 170 meV). Red spheres: Experimental data. Black dashed curve: Two-dimensional Wannier exciton model simultaneously fitting $\Delta\sigma_1$ and $\Delta\varepsilon_1$. $t_{PP} = 275$ fs, pump fluence, $\Phi = 16$ μJ/cm², lattice temperature $T_L = 300$ K.

fascinating class of objects in latest materials science. Their direct energy gaps in the optical range make them promising systems for novel optoelectronic applications[21]. Yet their optical properties are complicated by exotic room-temperature excitons, whose fundamental structure and dynamics has been under intense investigation[22]. While interband spectroscopy probes energies of excitons with vanishing centre-of-mass momenta, the majority of excitons have remained elusive, raising questions about their unusual internal structure, symmetry, many-body effects, and dynamics[23].

We utilize phase-locked multi-THz pulses to show the first direct experimental access to 1s excitons of all momenta in a single layer of WSe_2 (Ref. 7). Following resonant optical interband injection of 1s A excitons at a pump wavelength of 742 nm, a few-cycle multi-THz transient probes internal excitations of the quasiparticles as a function of the delay time (Fig. 1a). Electro-optic detection records amplitude and phase

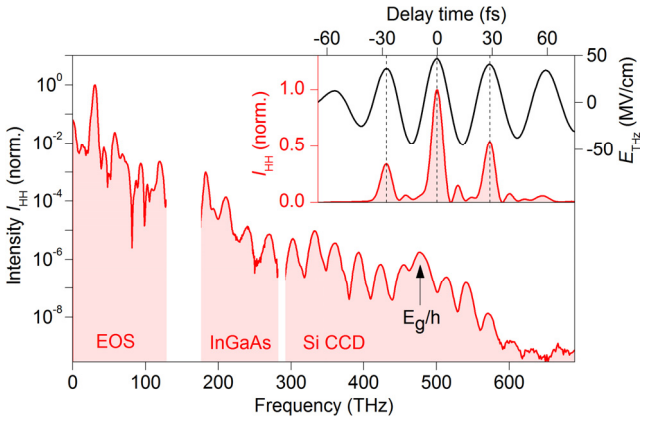

Fig. 2. Phase-locked THz high-harmonic generation in bulk GaSe. High-harmonic intensity spectrum (solid line and shaded area) emitted from a GaSe single crystal (thickness: 220 µm) driven by a phase-locked THz pulse centered at 30 THz. A combination of electro-optic sampling (EOS), an indium gallium arsenide diode array (InGaAs) and a silicon CCD (Si CCD) maps out the HH spectrum throughout the THz, far-infrared, mid-infrared, near-infrared, and visible regimes. E_g/h marks the bandgap frequency (476 THz). Inset: Temporal shape of intensity I_{HH} (red) of the high-harmonic pulse sequence relative to the driving multi-THz waveform (black). Note that the high-harmonic bursts are synchronized with the THz field maxima of positive polarity whereas high-harmonic emission is strongly suppressed for negative field crests.

of the transmitted oscillating electric field and allows us to retrieve the full complex dielectric function and its femtosecond evolution. The response functions clearly reveal the internal orbital 1s-2p resonance (Figs. 1b and c), which is highly sensitive to the shape of the excitonic envelope functions and provides accurate transition energies, oscillator strengths, densities and linewidths. Remarkably, the observed decay dynamics (inset of Fig. 1a) indicates a record fast radiative annihilation of small-momentum excitons within 150 fs, whereas Auger recombination prevails for optically dark states. The results provide a comprehensive view of excitons and introduce a new degree of freedom for quantum control, optoelectronics and valleytronics of dichalcogenide monolayers.

When quantum well excitons are embedded in high-quality optical microresonators, excitonic interband resonances can be strongly coupled with the trapped optical modes. The resulting light-matter mixed bosons, called exciton-polaritons feature an exceptionally low mass and have been shown to undergo a phase transition into a macroscopic quantum state often called a non-equilibrium Bose-Einstein condensate[24] (BEC). So far, however, only the photon component has been resolved, while the optically dark matter part has been a subject of controversial discussion. Sophisticated theories have suggested that the spontaneous quantum coherence is actually caused by ordinary photon lasing in the optical microcavities, while even the mere existence of excitons in the condensed regime has been challenged[25]. Here we trace the matter component of polariton condensates by monitoring intra-excitonic terahertz transitions[8]. We study how a reservoir of optically dark excitons forms and feeds the degenerate state. Intriguingly, the atom-like transition in excitons is dramatically renormalized upon macroscopic ground state population. Our results establish novel fundamental differences between polariton condensation and photon lasing and open unprecedented possibilities for quantum control of condensates.

Within the family of layered TMDCs, $TiSe_2$ has attracted special attention: Upon cooling below $T_c \approx 200$ K, it undergoes a transition into a commensurate charge-density wave (CDW) accompanied by the formation of a structural superlattice, the microscopic mechanisms of which have remained elusive. A first hypothesis assumes electron-phonon coupling based on a Jahn-Teller effect as the driving force[26]. A competing model suggests that Coulomb attraction may render the system unstable against the formation of excitons between the electron- and hole-like Fermi pockets[27], leading to lattice deformation with the corresponding wave vector. Here we show that ultrabroadband terahertz pulses can simultaneously trace the ultrafast evolution of coexisting lattice and electronic orders[9]. We demonstrate that the two components of the CDW order parameter – excitonic correlations and a periodic lattice distortion – respond vastly differently to 12-fs optical excitation. Even when the electronic order of the CDW is quenched, the lattice distortion can persist in a coherently excited state. The observation of this unprecedented phase proves that excitonic correlations are not the sole driving force of the CDW transition in 1*T*-$TiSe_2$, thus resolving a long-standing enigma. Disentangling strongly coupled components of order parameters in the time domain may facilitate a profound understanding of a broad class of phase transitions.

III. Lightwave-Driven Electronics in a Solid

Particle acceleration has been among the most successful concepts to reveal the structure and dynamics of matter. The recent achievement of intense, phase-locked few-cycle pulses in the ultrabroadband terahertz spectral range[15] allows this idea to be transferred to solid state physics by accelerating crystal electrons on a sub-cycle timescale[11-13,28]. Here, we drive electrons inside the semiconductor gallium selenide (GaSe) by applying a precisely adjustable high-field bias via phase-stable multi-THz pulses. The transient electric field reaching peak values of multiple 10 MV/cm simultaneously excites and accelerates electrons throughout the entire Brillouin zone of the solid[11]. These dynamics give rise to dynamical Bloch oscillations and the emission of a fully phase-stable high-order harmonic (HH) spectrum covering the entire THz-to-visible domain between 0.1 and 675 THz (Fig. 2). A surprising dependence of the emitted spectra on the carrier-envelope phase (CEP) of the driving waveform (not shown) hints at a delicate combination of inter- and intraband carrier dynamics[29] taking place on sub-cycle timescales.

Capturing the full quantum picture of HH generation in solids[30] requires a direct, time-resolved approach to study the high-field dynamics of electrons within a periodic crystal lattice on ultrafast timescales[31]. To this end, we introduce a novel cross-correlation scheme which enables the first time-domain observation of HH emission from a bulk solid with direct temporal correlation to the driving field[13]: HHs are emitted as a sequence of ultrashort, nearly unchirped bursts,

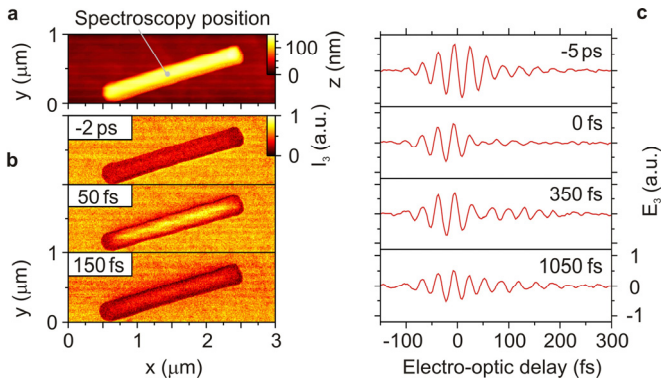

Fig. 3. Pump-probe multi-THz spectroscopy and microscopy of a single InAs nanowire. **a**, Topography of the InAs nanowire recorded by AFM. **b**, Scattered intensity I_3 of the multi-THz probe pulse as a function of near-IR pump/multi-THz probe delay time and AFM tip position. **c**, Scattered electric near-field (E_3) waveforms recorded at the spectroscopy position denoted in **a** at different times after photoexcitation. The oscillating electric near field from a $(10\ \text{nm})^3$ volume is traced with 10 fs temporal resolution.

locked to the driving field maxima of one polarity only (inset of Fig. 2). This structure is the hallmark of a quantum interference of several interband excitation pathways including strong-field transitions between initially Pauli-blocked valence bands. A subtle frequency modulation within single emission events reflects signatures of coherent intraband transport, as expected from the Bloch acceleration of charge carriers[32]. The direct observation of lightwave-controlled electron dynamics marks the way towards a complete microscopic picture of HHG in solids, ultrafast electronics, all-optical band structure reconstruction, and novel solid-state CEP-stable attosecond sources[33].

IV. Field-Sensitive Near-Field Microscopy

While ultrashort THz transients offer efficient ways to observe and control low-energy excitations on the femtosecond scale, conventional THz spectroscopy is intrinsically restricted to ensemble measurements by the diffraction limit. As a result, it probes dielectric functions averaged over the size, structure, orientation and density of nanoparticles, nanocrystals or nanodomains. Ultrafast THz spectroscopy studies beyond the diffraction limit have been a longstanding goal.

It has been demonstrated that coupling THz pulses to sharp metallic tips encodes subwavelength spatial information onto the scattered fields, which have been detected either by intensity-resolving measurements in the multi-THz range[18,34] or electro-optic sampling in the few-THz window[17]. None of these studies, however, has probed photoinduced dynamics in single particles with nanometer lateral dimensions. Conversely, time-integrated intensity detection has been used to measure multi-THz pulses scattered from photoexcited graphene with a temporal resolution of 200 fs, limited by the duration of the THz probe pulse[35]. Following a different scheme, ultrafast charging dynamics in single nanoparticles have recently been measured electronically on the 1 nm scale using terahertz scanning tunnelling microscopy (THz-STM), where local currents are induced by few-THz field transients[16].

Here, we use electro-optic sampling of multi-THz pulses to directly trace the scattered electric near field with a 10-fs gate pulse, revealing the dynamics of the dielectric function at a nanowire surface with 10-nm spatial resolution[14]. Our experiment marks the first field-resolved pump-probe spectroscopy on the nanoscale and introduces sub-nanoparticle spatial resolution to sub-cycle multi-THz studies. Additionally, by resolving the time-dependent dielectric function, we establish a complementary approach to transport measurements by THz-STM, while providing access to new non-metallic systems and faster dynamics.

V. Conclusions

The possibility to shape electromagnetic fields with sub-cycle and nanometer precision and to reach atomic field strengths opens up spectacular opportunities to control photons, charge and spin at optical clock rates. While so far we have shown lightwave-driven electronics and near-field probing in separate experiments, we anticipate that the combination of strong-field acceleration with a scanning near-field microscope will ultimately pave the way towards complete four-dimensional control of the microscopic constituents of matter on atomic length and time scales.

References

[1]. M. Tonouchi, "Cutting-edge terahertz technology", *Nature Photon.* 1, 97 (2007).

[2]. R. Ulbricht, E. Hendry, J. Shan, T. F. Heinz, and M. Bonn, "Carrier dynamics in semiconductors studied with time-resolved terahertz spectroscopy", *Rev. Mod. Phys.* 83, 543 (2011).

[3]. P. U. Jepsen, D. G. Cooke, and M. Koch, "Terahertz spectroscopy and imaging – modern techniques and applications", *Laser Photon. Rev.* 5, 124 (2011).

[4]. T. Kampfrath, K. Tanaka, and K. A. Nelson, "Resonant and nonresonant control over matter and light by intense terahertz transients", *Nature Photon.* 7, 680 (2013).

[5]. G. Günter, A. A. Anappara, J. Hees, A. Sell, G. Biasiol, L. Sorba, S. De Liberato, C. Ciuti, A. Tredicucci, A. Leitenstorfer, and R. Huber, "Sub-cycle switch-on of ultrastrong light-matter interaction", *Nature* 458, 178 (2009).

[6]. T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, "Coherent terahertz control of antiferromagnetic spin waves", *Nature Photon.* 5, 31 (2011).

[7]. C. Poellmann, P. Steinleitner, U. Leierseder, P. Nagler, G. Plechinger, M. Porer, R. Bratschitsch, C. Schüller, T. Korn, and R. Huber, "Resonant internal quantum transitions and femtosecond radiative decay of excitons in monolayer WSe_2", *in press.*

[8]. J.-M. Ménard, C. Poellmann, M. Porer, U. Leierseder, E. Galopin, A. Lemaître, A. Amo, J. Bloch, and R. Huber, "Revealing the dark side of a bright exciton-polariton condensate", *Nature Commun.* 5, 4648 (2014).

[9]. M. Porer, U. Leierseder, J.-M. Ménard, H. Dachraoui, L. Mouchliadis, I. E. Perakis, U. Heinzmann, J. Demsar, K. Rossnagel, and R. Huber, "Non-thermal separation of electronic and structural orders in a persisting charge density wave", *Nature Mater.* 13, 857 (2014).

[10]. K. W. Kim, A. Pashkin, H. Schäfer, M. Beyer, M. Porer, T. Wolf, C. Bernhard, J. Demsar, R. Huber, and A. Leitenstorfer, "Ultrafast transient generation of spin-density-wave order in the normal state of $BaFe_2As_2$ driven by coherent lattice vibrations", *Nature Mater.* 11, 497 (2012).

[11]. O. Schubert, M. Hohenleutner, F. Langer, B. Urbanek, C. Lange, U. Huttner, D. Golde, T. Meier, M. Kira, S. W. Koch and R. Huber, "Sub-cycle control of terahertz high-harmonic generation by dynamical Bloch oscillations", *Nature Photon.* 8, 119 (2014).

[12]. C. Lange, T. Maag, M. Hohenleutner, S. Baierl, O. Schubert, E. Edwards, D. Bougeard, G. Woltersdorf, and R. Huber, "Extremely

nonperturbative nonlinearities in GaAs driven by atomically strong terahertz fields in gold metamaterials", *Phys. Rev. Lett.* 113, 227401 (2014).
[13]. M. Hohenleutner, F. Langer, O. Schubert, M. Knorr, U. Huttner, S. W. Koch, M. Kira, and R. Huber, "Real-time observation of interfering crystal electrons in high-harmonic generation", *in press.*
[14]. M. Eisele, T. L. Cocker, M. A. Huber, M. Plankl, L. Viti, D. Ercolani, L. Sorba, M. S. Vitiello, and R. Huber, "Ultrafast multi-terahertz nano-spectroscopy with sub-cycle temporal resolution", *Nature Photon.* 8, 841 (2014).
[15]. A. Sell, A. Leitenstorfer, and R. Huber, "Phase-locked generation and field-resolved detection of widely tunable terahertz pulses with amplitudes exceeding 100 MV/cm", *Opt. Lett.* 33, 2767 (2008).
[16]. T. L. Cocker, V. Jelic, M. Gupta, S. J. Molesky, J. A. J. Burgess, G. De Los Reyes, L. V. Titova, Y. Y Tsui, M. R. Freeman, and F. A. Hegmann, "An ultrafast terahertz scanning tunnelling microscope", *Nature Photon*. **7**, 620 (2013).
[17]. K. Moon, Y. Do, M. Lim, G. Lee, H. Kang, K.-S. Park, and H. Han, "Quantitative coherent scattering spectra in apertureless terahertz pulse near-field microscopes", *Appl. Phys. Lett*. **101**, 011109 (2012).
[18]. R. Jacob, S. Winnerl, M. Fehrenbacher, J. Bhattacharyya, H. Schneider, M. T. Wenzel, H.-G. von Ribbeck, L. M. Eng, P. Atkinson, O. G. Schmidt, and M. Helm, "Intersublevel spectroscopy on single InAs-quantum dots by terahertz near-field microscopy", *Nano Lett.* 12, 4336 (2012).
[19]. M. Porer, J.-M. Ménard, and R. Huber, "Shot noise reduced terahertz detection via spectrally postfiltered electro-optic sampling", *Opt. Lett.* 39, 2435 (2014).
[20]. O. Schubert, C. Riek, F. Junginger, A. Sell, A. Leitenstorfer, and R. Huber, "Ultrashort pulse characterization with a terahertz streak camera", *Opt. Lett.* 36, 4458 (2011).
[21]. F. H. L. Koppens, T. Mueller, Ph. Avouris, A. C. Ferrari, M. S. Vitiello, and M. Polini, "Photodetectors based on graphene, other two-dimensional materials and hybrid systems", *Nature Nanotechnol. 9*, 780 (2014).
[22]. A. Chernikov, T. C. Berkelbach, H. M. Hill, A. Rigosi, Y. Li, O. Burak Aslan, D. R. Reichman, M. S. Hybertsen, and T. F. Heinz, "Exciton Binding Energy and Nonhydrogenic Rydberg Series in Monolayer WS_2", *Phys. Rev. Lett.* 113, 076802 (2014).
[23]. Z. Ye, T. Cao, K. O'Brien, H. Zhu, X. Yin, Y. Wang, S. G. Louie, and X. Zhang, "Probing excitonic dark states in single-layer tungsten disulphide", *Nature* 513, 214 (2014).
[24]. J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymanska, R. André, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, and Le Si Dang, "Bose–Einstein condensation of exciton polaritons", *Nature* 443, 409-414 (2006).
[25]. H. M. Gibbs, G. Khitrova, and S. W. Koch, "Exciton–polariton light–semiconductor coupling effects". *Nature Photon.* 5, 273–282 (2011).
[26]. T. E. Kidd, T. Miller, M. Y. Chou, and T.-C. Chiang, "Electron–hole coupling and the charge density wave transition in $TiSe_2$", *Phys. Rev. Lett.* 88, 226402 (2002).
[27]. H. Cercellier, C. Monney, F. Clerc, C. Battaglia, L. Despont, M. G. Garnier, H. Beck, P. Aebi, L. Patthey, H. Berger, and L. Forró, "Evidence for an excitonic insulator phase in 1T-$TiSe_2$", *Phys. Rev. Lett.* 99, 146403 (2007).
[28]. B. Zaks, R. B. Liu, and M. S. Sherwin, "Experimental observation of electron-hole recolissions", *Nature* 483, 580 (2012).
[29]. D. Golde, M. Kira, T. Meier, and S. W. Koch, "Microscopic theory of the extremely nonlinear terahertz response of semiconductors", *Phys. Status Solidi B* 248, 863 (2008).
[30]. S. Ghimire, A. D. DiChiara, E. Sistrunk, P. Agostini, L. F. DiMauro, and D. A. Reis, "Observation of high-order harmonic generation in a bulk crystal", *Nature Phys.* 7, 138 (2011).
[31]. T. Higuchi, M. I. Stockman, and P. Hommelhoff, "Strong-Field Perspective on High-Harmonic Radiation from Bulk Solids", *Phys. Rev. Lett.* 113, 213901 (2014).
[32]. C. A. Zener, "Theory of the Electrical Breakdown of Solid Dielectrics", *Proc. R. Soc. Lond. A* 145, 523 (1934).
[33]. F. Krausz, and M. I. Stockman, "Attosecond metrology: from electron capture to future signal processing", *Nature Photon.* 8, 205 (2014).
[34]. J. M. Stiegler, A. J. Huber, S. L. Diedenhofen, J. Gómez Rivas, R. E. Algra, E. P. A. M. Bakkers, and R. Hillenbrand, "Nanoscale free-carrier profiling of individual semiconductor nanowires by infrared near-field nanoscopy", *Nano Lett*. **10**, 1387 (2010).
[35]. M. Wagner, Z. Fei, A. S. McLeod, A. S. Rodin, W. Bao, E. G. Iwinski, Z. Zhao, M. Goldflam, M. Liu, G. Dominguez, M. Thiemens, M. M. Fogler, A. H. Castro Neto, C. N. Lau, S. Amarie, F. Keilmann, and D. N. Basov, "Ultrafast and nanoscale plasmonic phenomena in exfoliated graphene revealed by infrared pump-probe nanoscopy" *Nano Lett*. **14**, 894 (2014).