

Development and Application of Ultrafast Transmission Electron Microscope Based on Schottky Field Emission

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In recent years, we have witnessed the substantial progress of ultrafast transmission electron microscopy (UTEM) [1,2]. As a powerful table-top apparatus, UTEM is challenging the resolution abilities of our instrument in both spatial and temporal domain, thanks to the well-developed pump-probe and electron microscopy techniques [3,4]. With its nanometer and sub-picosecond spatiotemporal resolution capabilities, UTEM has enabled the visualization of numerous important transient processes, including the lattice relaxation, the phase transition [5], the nanomechanical motions, the spin [6] and electronic dynamics [7-9] for nanoscale materials or structures ranging from physics to chemistry and biology. In particular, UTEM also lead to a research direction completely outside the realm of conventional TEM, i.e. photon-induced-near-field electron microscopy (PINEM), an unique experimental technique that can image the light-electron interactions near nanostructures or at an interface, providing exciting prospects for the investigation of dynamics of photonics and plasmonics [10].

In our laboratory, an ultrafast transmission electron microscope (UTEM) has been developed based on Schottky-type field emission gun (FEG) (Institute of Physics, Chinese Academy of Sciences). This setup can operate either in continuous or pulsed mode, and the electron pulse emission is achieved by integrating a laser port between the electron gun and the column. For pulsed mode, the optimized electron beam properties are of ~ 0.65 eV energy monochromaticity, micrometer-scale coherence lengths and ~ 300 fs pulse duration. Then these UTEMs have been extensively improved for photoemission imaging and time-resolved observations, including ultrafast electron diffraction/imaging, Lorentz UTEM, time-resolved electron energy-loss spectroscopy (EELS) and photoinduced near-field electron microscopy (PINEM) [11], as shown in Fig.1.

Recently, we have performed a variety of investigations on the structural dynamics of functional materials in our laboratory. For instance, the notable anisotropic lattice dynamics in multi-walled C- and BN-nanotubes have been studied in a full reversible cycle with a time scale from picoseconds to hundreds of microseconds, moreover, a visible change of energy band-gap was found on the BN-nanotubes after a fs-laser excitation[12], as shown in Fig.2. The martensitic transition in the Heusler alloy $Mn_{50}Ni_{40}Sn_{10}$ has also been investigated using the time-resolved imaging and ultrafast diffraction[13]. The photoinduced magnetic processes have been directly observed using time-resolved Lorentz TEM imaging. Very recently, the nonequilibrium phase transition between CDW states and the relevant hidden quantum states has been carefully investigated and analyzed in low-dimensional system $1T-TaS_{2-x}Se_x$ at low temperatures [14, 15, 16].

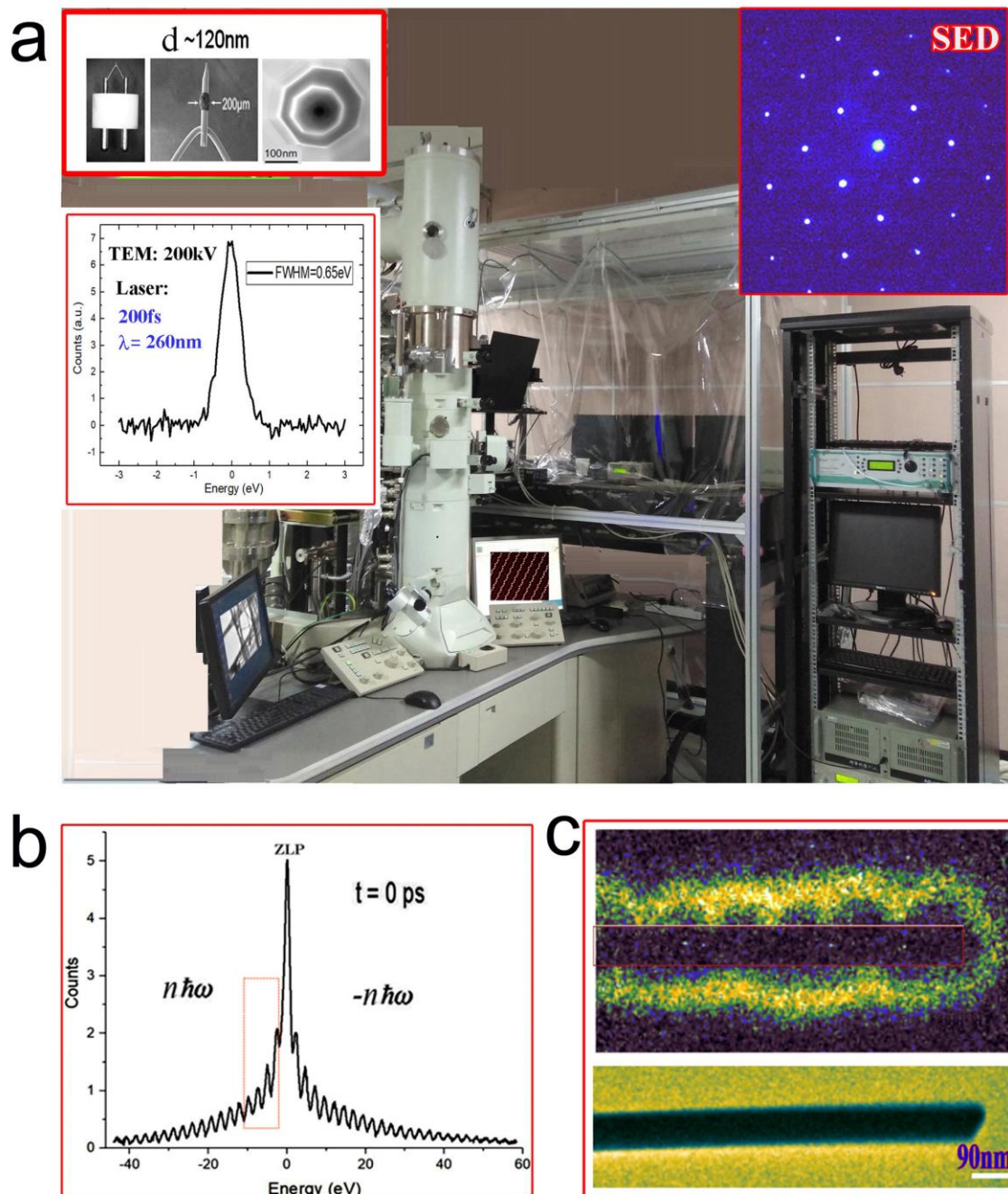


Figure 1. The UTEM apparatus based on a Schottky-type field emission gun and its performance in photo-emission mode. (a) A photograph of UTEM and the fs-laser instruments in our laboratory. The inserted figures show images and data of Schottky-field emitter, the EELS spectrum with energy resolution of 0.65eV and an electron diffraction pattern of the Au-crystal taken from pulsed mode. (b) EELS data taken at the delay time of $t=0$ for an Ag nanorod irradiated with a fs-laser pulse. (c) Relevant PINEM image obtained for an individual nanorod ($\sim 90\text{nm}$) (exposure time $\sim 100\text{ s}$), and a convention TEM image is also shown for comparison.

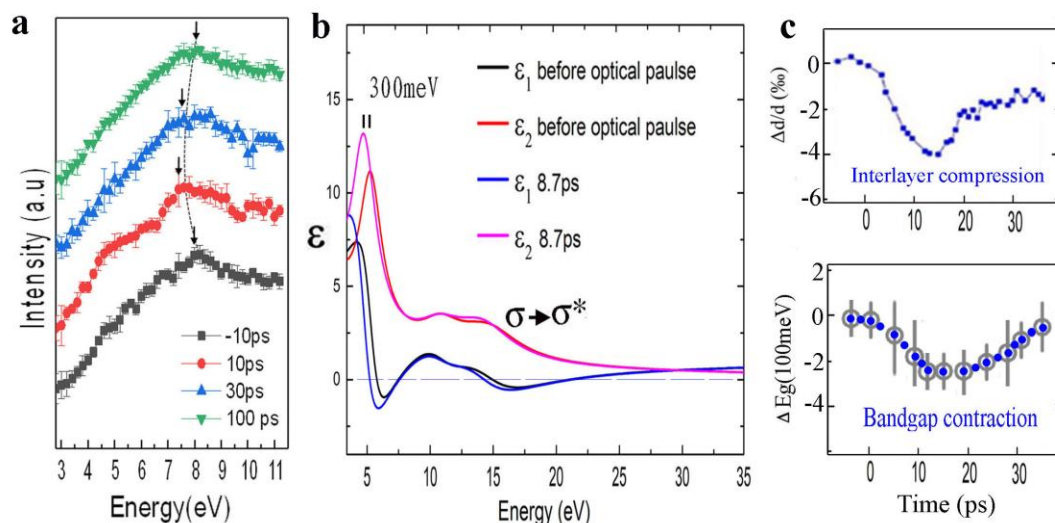


Figure 2. Time-resolved analysis of electronic structural changes in BN nanotubes. (a) The plasmon peaks shift with the time delays, illustrating the valence excitation features of the inter band transitions upon fs-laser excitation. (b) Dielectric function of the BNNTs showing the real and imaginary parts derived from the loss function, which yields a clear contraction of the energy bandgap at an excited state. (c) Non-thermal change in the interlayer spacing and contraction of the energy bandgap as measured from -10 ps to 40 ps, showing the cooperative evolution of the bandgap and the nonthermal structural transient in BN nanotubes.

References

- [1] A.H. Zewail, et al *Science* 328 (2010) 187-193.
- [2] N.D. Browning, et al, *Chem. Phys. Chem.* 11 (2010) 781-782.
- [3] P. Baum, et al, *PNAS* 104 (2007) 18409-18414.
- [4] Y. Morimoto, et al, *Nat. Phys.* 14 (2018) 252-256.
- [5] S.R.M. van der Veen, et al, *Nat. Chem.* 5 (2013) 395-402.
- [6] G. Berruto, et al, *Phys. Rev. Lett.* 120 (2018) 117201.
- [7] F. Carbone, et al, *Chem. Phys. Lett.* 468 (2009) 107-111.
- [8] L. Piazza, et al, *Struct. Dyn.* 1, (2014) 014501.
- [9] F. Carbone, et al, *Struct. Dyn.* 2 (2015) 020601.
- [10] B. Barwick, et al, *Nature* 462 (2009) 902-906.
- [11] C.H. Zhu, D.G. Zheng, et al, *Ultramicroscopy* 209, 112887 (2020).
- [12] Z. W. Li, R.J. Xiao, et al, *ACS Nano* 13, 11623 (2019); Z. W. Li, S. Sun, et al., *Nanoscale*, **9**, 13313 (2017).
- [13] M. Zhang, G. Cao, H. Tian, et al., *Physical Review B*, **96**, 174203 (2017).
- [14] K. Sun, S. Sun, C. Zhu A. et al., *Science Advances*, **4**, eaas9660 (2018).
- [15] H. Yang, S. Song, M. Zhang, et al., *Chin. Phys. B*, **27**, 70703 (2018).
- [16] This work was supported by the National Key Research and Development Program of China under Grant Nos. 2016YFA0300303, 2017YFA0504703, and 2017YFA0302904, 2017YFA0303000, the National Basic Research Program of China under Grant No. 2015CB921304.