Review of Scientific Instruments

Development of *in situ* optical spectroscopy with high temporal resolution in an aberration-corrected transmission electron microscope

Cite as: Rev. Sci. Instrum. **92**, 013704 (2021); https://doi.org/10.1063/5.0031115
Submitted: 29 September 2020 . Accepted: 01 January 2021 . Published Online: 27 January 2021

Chang Liu, Chaojie Ma, Jinjing Xu, Ruixi Qiao, Huacong Sun, Xiaomin Li, Zhi Xu, Peng Gao, Enge Wang, Kaihui Liu, and Xuedong Bai

Review of Scientific Instruments ARTICLE scitation.org/journal/rsi

Development of *in situ* optical spectroscopy with high temporal resolution in an aberration-corrected transmission electron microscope

Cite as: Rev. Sci. Instrum. 92, 013704 (2021); doi: 10.1063/5.0031115
Submitted: 29 September 2020 • Accepted: 1 January 2021 •
Published Online: 27 January 2021

Chang Liu,[1] Chaojie Ma,[1] Jinjing Xu,[2] Ruixi Qiao,[1] Huacong Sun,[2] Xiaomin Li,[2] Zhi Xu,[2] Peng Gao,[1] Enge Wang,[1] Kaihui Liu,[1,a)] and Xuedong Bai[2,a)]

AFFILIATIONS

[1] School of Physics, Peking University, Beijing 100871, China
[2] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

[a)] **Authors to whom correspondence should be addressed:** khliu@pku.edu.cn and xdbai@iphy.ac.cn

ABSTRACT

Exploring the corresponding relation between structural and physical properties of materials at the atomic scale remains the fundamental problem in science. With the development of the aberration-corrected transmission electron microscopy (AC-TEM) and the ultrafast optical spectroscopy technique, sub-angstrom-scale spatial resolution and femtosecond-scale temporal resolution can be achieved, respectively. However, the attempt to combine both their advantages is still a great challenge. Here, we develop *in situ* optical spectroscopy with high temporal resolution in AC-TEM by utilizing a self-designed and manufactured TEM specimen holder, which has the capacity of sub-angstrom-scale spatial resolution and femtosecond-scale temporal resolution. The key and unique design of our apparatus is the use of the fiber bundle, which enables the delivery of focused pulse beams into TEM and collection of optical response simultaneously. The generated focused spot has a size less than 2 μm and can be scanned in plane with an area larger than 75 × 75 μm^2. Most importantly, the positive group-velocity dispersion caused by glass fiber is compensated by a pair of diffraction gratings, thus resulting in the generation of pulse beams with a pulse width of about 300 fs (@ 3 mW) in TEM. The *in situ* experiment, observing the atomic structure of CdSe/ZnS quantum dots in AC-TEM and obtaining the photoluminescence lifetime (~4.3 ns) in the meantime, has been realized. Further ultrafast optical spectroscopy with femtosecond-scale temporal resolution could be performed in TEM by utilizing this apparatus.

I. INTRODUCTION

Transmission electron microscopy (TEM) is a versatile tool for material characterization, providing structural and electronic information of materials by imaging, electron diffraction, and spectroscopy.[1–4] In recent decades, with the development of aberration correction technology, sub-angstrom spatial resolution has been achieved in aberration-corrected TEM (AC-TEM), which promotes great progress in condensed-matter physics and materials science.[5–17] In the meantime, with the desire of exploring the influence of external stimuli to the specimen in real time, *in situ* TEM is another developing field in modern electron microscopy.[18,19] The combination of AC-TEM and *in situ* TEM technology has provided an unparalleled capability to study the relationship between the properties and structure of materials with atomic-scale spatial resolution.[20–22] Until now, through ingeniously designed specimen holders, a variety of external stimuli has been brought into TEM, such as mechanical force,[23–25] electric field,[26–30] heat,[31,32] and light.[33–46] One of the pioneer methods for *in situ* light illumination is achieved through an optical fiber or some optical elements.[33–37]

However, these implementations are complicated and often need the modification of the TEM column, such as inserting some optical windows and parabolic mirrors. In fact, lots of efforts have been taken to introduce optical signals into TEM, including *in situ* characterization of photoluminescence (PL) spectroscopy,[34] Raman spectroscopy,[37,38] laser-induced phase transformation,[41,43,44] and photocatalysts.[45,46] Despite this, the excitation efficiency is still low due to the limitation of the power density of the radiation light. Moreover, it is still a challenge in TEM to achieve *in situ* optical spectroscopy with high temporal resolution, which uses ultrashort pulse lasers to study dynamics on extremely short time scales, including attosecond-to-picosecond spectroscopy (such as ultrafast transient absorption using a pump-probe scheme) and picosecond-to-nanosecond spectroscopy [such as using a streak camera and time-correlated single photon counting (TCSPC)].[47]

In this work, we have successfully designed and manufactured a distinctive specimen holder, in which the optical part consists of a fiber bundle and a 4*f* lens system. With the assistance of the outer optical components, focused laser beams with a spot size less than 2 μm can be introduced into TEM through the fiber bundle. The focused spot can be scanned in an area larger than 75 × 75 μm^2 with a resolution of 10 nm. More impressively, pulse laser beams with a pulse width of about 300 fs (@ 3 mW) can be focused on the sample, enabling *in situ* optical measurement with high temporal resolution in TEM. We have observed the atomic structure of CdSe/ZnS quantum dots and utilized TCSPC to *in situ* measure the PL lifetime in the AC-TEM specimen chamber. Further ultrafast optical spectroscopy such as pump-probe measurement could be achieved by utilizing our self-designed specimen holder with a minor modification, which will be discussed in the following part.

II. EXPERIMENTAL SETUP

In order to introduce laser into AC-TEM (JEM-ARM 300F, JEOL), a fiber bundle is inserted and fixed at the center of the specimen holder. With the help of a 4*f* de-magnified system composed of two lenses installed on the holder, a focused laser spot can be finally introduced into the specimen chamber. The schematics for *in situ* optical spectroscopy with high temporal resolution in AC-TEM are shown in Fig. 1(a). The fiber bundle consisting of 5600 single-mode or few-mode (@800 nm) cores is bought from Schott (Schott, 1 639 903), with a length of 85 cm and a core-to-core distance of 6.9 μm. The femtosecond pulse laser (~130 fs) generated by a mode locked Ti:sapphire laser with a 76 MHz repetition rate is employed as the laser source. The emitted femtosecond pulse laser with a center wavelength of 800 nm is first expanded and collimated. Then, to compensate the positive group-velocity dispersion caused by the glass fiber, the collimated pulse beam passes through a dispersion compensation module, consisting of a pair of diffraction gratings and a mirror M. The pulse width can be compressed to hundreds of femtoseconds, which guarantees the temporal resolution for ultrafast optical spectroscopy. The realization process of pulse compression will be explained in detail in the following part. Then, a microscope object (MO) is used to focus the pulse beam on the proximal facet of the fiber bundle, which is mounted on a three-dimensional translational stage. The distal end of the fiber bundle is installed into the TEM specimen holder. Together with a 4*f* de-magnified system composed of two lenses (focal lengths of L1 = 15 mm and L2 = 7.5 mm) with a diameter of 6.25 mm, the output pulse beam is finally guided into TEM with a size less than 2 μm. By choosing different input cores, the position of output focus will change accordingly, thus achieving focus scanning. To fill the spacing between neighbor cores, a two-dimensional piezo stage is used to shift the focus position more precisely with a resolution of 10 nm. As the pulse beam is focused on the sample, the optical response of the sample is collected by the same fiber bundle, which will be led to an image sensor or spectrometer. A single-photon avalanche photodiode (SPAD) detector equipped with a TCSPC module is used to collect the time-resolved PL signal. Our apparatus is powerful to perform *in situ* time-resolved optical measurement using pulse laser, such as the measurement of PL lifetime. The atomic structure of CdSe/ZnS quantum dots is characterized and its PL lifetime is measured *in situ* using TCSPC. Based on our apparatus, further *in situ* ultrafast optical spectroscopy such as pump-probe measurement in TEM is promising, which will unveil an ultrafast dynamics nature of materials with atomic-scale spatial resolution.

III. SPECIMEN HOLDER AND EXPERIMENTAL DETAILS

A. Design details of the specimen holder

The self-designed and manufactured specimen holder mainly consists of five parts [Figs. 1(b) and 2], including the main body of the holder, the fiber bundle, a 4*f* de-magnified system, translational stages, and a motion control system. The main body of the holder is made of aluminum, and a tungsten tip is located at the front part. A 4*f* de-magnified system composed of two lenses, L1 and L2, with a diameter of 6.25 mm is fixed near the tip. The 4*f* system is used to deliver the pulse beam into the TEM column and form a confocal system. The fiber bundle is inserted in the center of the main body of the specimen holder, and the distal end is fixed near the back focal plane of lens L1. There is a two-dimensional piezo stage located at the distal end of the fiber bundle to shift the laser spot precisely with a resolution of 20 nm, which means a resolution of 10 nm for the focal spot scanning because of the 4*f* de-magnified system. In order to guide the input laser into the TEM, the proximal end of the fiber bundle reaches out of the holder and is mounted on a three-dimensional translational stage to optimize the laser coupling and scan the focal spot in the *x*–*y* plane. The distance between the distal facet and lens L1 can be adjusted in the axial direction by a one-dimensional translational stage. The motion control system provides power to drive the piezo stage via several electric cables inserted into the specimen holder beforehand.

B. Laser focusing and scanning

Due to the space limitation of the pole pieces, it is difficult to observe and locate the generated focal spot in TEM. Therefore, a white light imaging module is adopted for *in situ* laser focusing. In this module, the white light from a halogen lamp source is first collimated by lens L4 and then passes through the beam splitters BS2 and BS1. The white light is focused to illuminate the whole proximal facet of the fiber bundle by the MO. Both the reflected white light and pulse beam from the proximal facet are collected by the same MO. The collected signals pass through BS1 and BS2

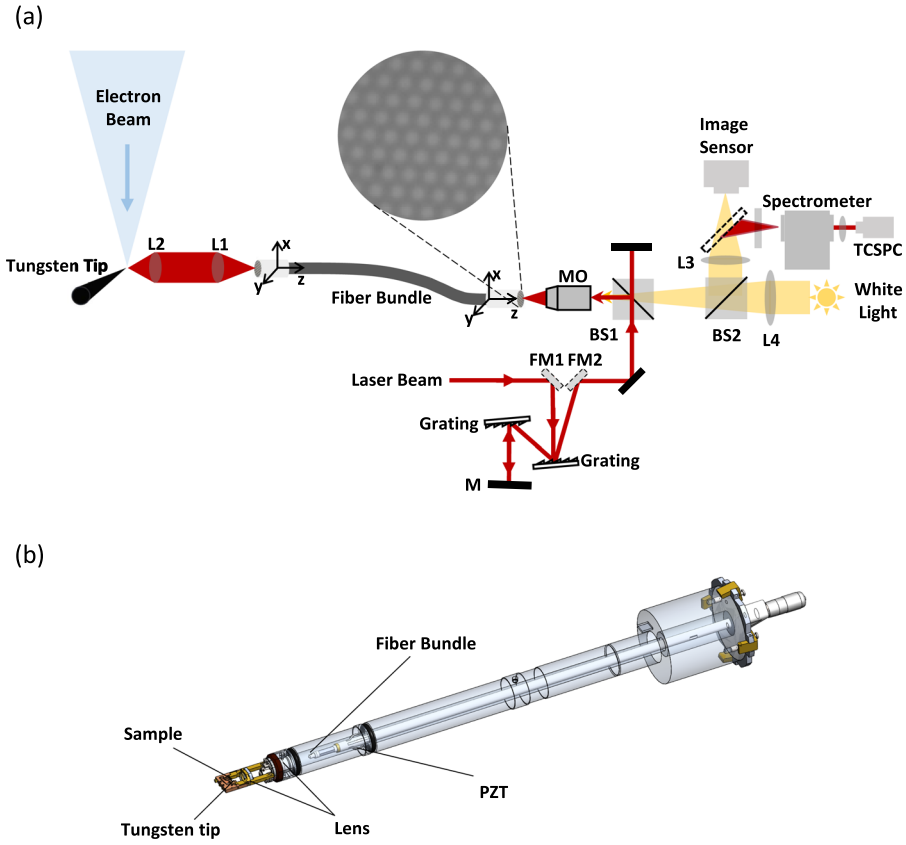

FIG. 1. Schematic representation of the experimental setup: (a) schematics for *in situ* optical spectroscopy with high temporal resolution in AC-TEM. MO: microscope objective; L1, L2, L3, and L4: lenses; BS1 and BS2: beam splitters; M: mirror; FM1 and FM2: flip mirrors; and TCSPC: time-correlated single photon counting module and (b) the schematic design of the specimen holder.

and are imaged on the image sensor via lens L3. According to the image of the proximal facet [Fig. 3(a)], the pulse beam could be accurately coupled into a certain single core, thus achieving *in situ* focusing.

To characterize the quality of the focused spot, the front end of the specimen holder is taken down intentionally outside of the TEM. The focused pulse beam is collected and magnified by a long working distance microscope objective and imaged on an image sensor target via a tube lens (not given in the schematics). After passing through the 4*f* de-magnified system, a focal spot with a size of ~1.7 μm is achieved [Figs. 3(b) and 3(c)]. In principle, the focal spot could be further reduced by using a lens with a shorter focal length. However, the resulting closer distance between the lens and the tungsten tip would potentially influence the electron beam in TEM and has the risk of damaging the optical components. By adjusting the proximal end position in the *x*–*y* plane to couple laser into different cores, *in situ* focus scanning could be achieved in TEM, with a scanning range larger than 75 μm in any direction in the *x*–*y* plane [Fig. 3(d)]. In order to fill the spacing between the neighbor cores to achieve high-precision scanning, the distal end of the fiber can be shifted by a two-dimensional translational piezo stage to achieve focal spot scanning with a resolution of 10 nm. The focus could be scanned more precisely in the *x*–*y* plane by using the piezo stage [Fig. 3(e)].

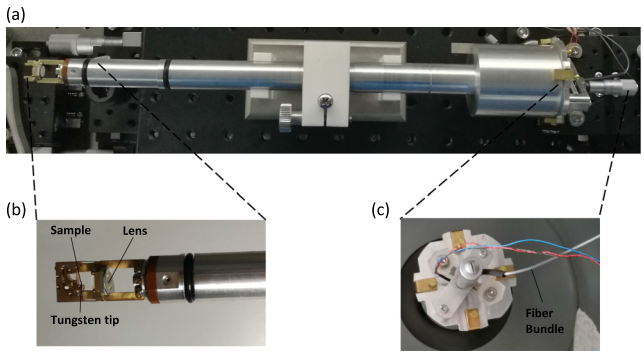

FIG. 2. Photographs of the specimen holder showing (a) the main body, (b) the front end of the holder containing the lens and tungsten tip, and (c) the back end of the holder. The fiber bundle is inserted into the middle of the holder.

C. Pulse compression

As mentioned above, when passing through a normal glass fiber, the pulses will be broadened, resulting from dispersion and nonlinearities of the fiber [Fig. 4(a)].[48–54] In our experiment, an 85 cm-long fiber bundle is used, and the femtosecond pulse is broadened to a picosecond pulse after passing through the fiber

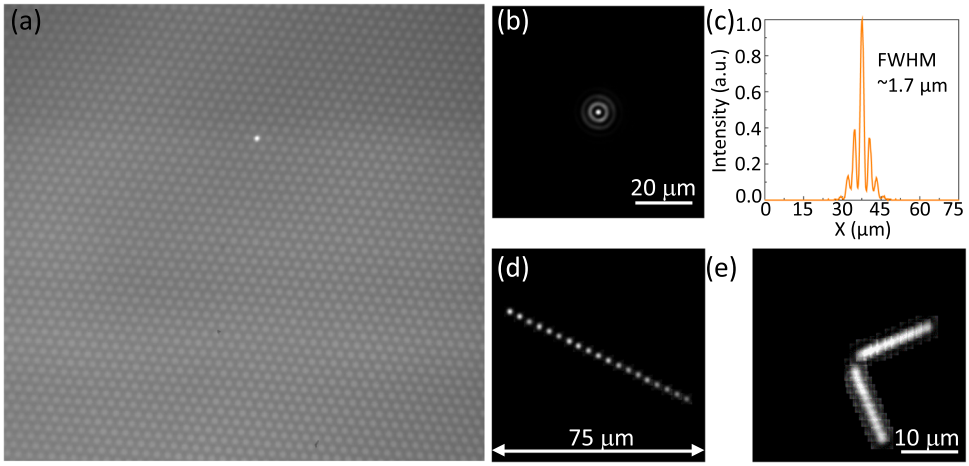

FIG. 3. *In situ* focusing and scanning using the self-designed specimen holder: (a) the optical image of the proximal facet of the fiber bundle, clearly showing the cores of the fiber bundle and the laser spot, (b) the generated focal spot after passing through the 4*f* de-magnified system, (c) the intensity profile of the focal spot along the horizontal direction, (d) *in situ* focus scanning in the *x*–*y* plane showing a scanning area larger than 75 × 75 μm^2, and (e) high-precision scanning of the focus realized by the piezo stage.

[Fig. 4(b)]. In order to compensate the positive group velocity dispersion (GVD) caused by the fiber, the collimated pulse beam passes through a dispersion compensation module,[49,50,53,55,56] as shown in Fig. 1. In this module, one of the diffraction gratings is mounted on a one-dimensional translational stage to adjust the distance between the grating pair. After passing through the grating pair doubly, the pulse beam is negatively chirped. The propagation direction of the pre-chirped pulse beam reflected by a flip mirror FM2 is the same as that without the flip mirrors FM1 and FM2 in the system. Consequently, the negatively pre-chirped pulse beam will be positively chirped through the fiber bundle, and the final output pulse can nearly maintain the original pulse width [Fig. 4(c)]. The compensated pulse width is measured to be about 300 fs [Fig. 4(d)] by the commercial auto-correlator at an output power of 3 mW. It is worth noting that with the increase in the input laser power, the nonlinear effects of the fiber such as self-phase modulation become more significant, and the resulting broadening cannot be fully compensated by negative pre-chirping in our experiment.[49,50] The generation of compressed pulses enables us to conduct time-resolved experiments and even ultrafast optical spectroscopy in TEM with hundred-femtosecond temporal resolution.

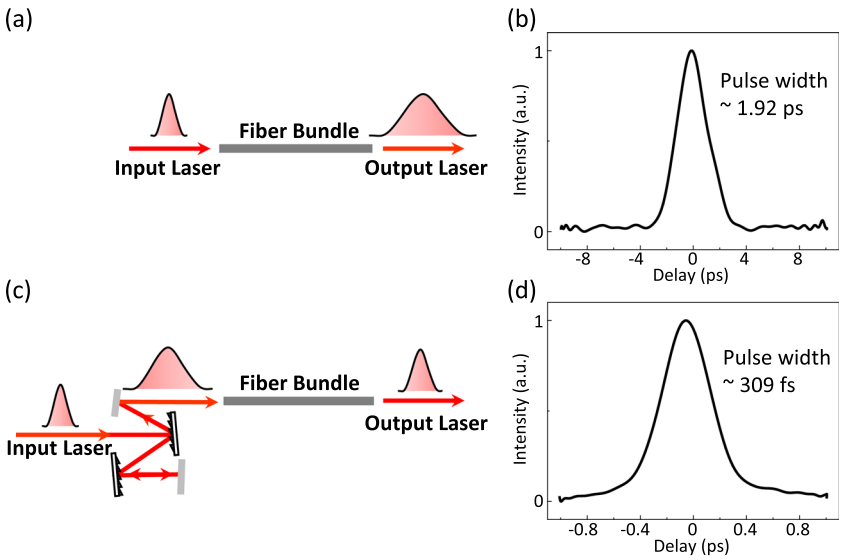

FIG. 4. Pulse broadening and compression: (a) the pulse is broadened after passing through the fiber bundle caused by positive GVD of the fiber, (b) the resulting pulse width of the output pulse beam without the dispersion compensation module, (c) the process of pulse compression using the diffraction grating pair, and (d) the compensated pulse width of the output pulse beam with the use of the dispersion compensation module.

IV. RESULTS AND DISCUSSIONS

A. *In situ* measurement of PL lifetime in AC-TEM

In situ time-resolved optical measurement in AC-TEM is feasible by utilizing our specimen holder. As for sample preparation, first, we prepared the tungsten tip with a proper length by electrochemical corrosion and dropped the colloidal quantum dots (CdSe/ZnS) onto the tip top under an optical microscope. Then, the tip was installed at the front end of the sample holder, and the holder is inserted into the AC-TEM specimen chamber for *in situ* measurement. In order to make the sample into the area of the focal spot, we chose PL signals from the sample as feedback. The focused laser spot was introduced, accompanied with PL signal collection. Then, the focused spot was moved by adjusting the coupling core and the axial position of the distal facet. With the maximal PL signal collected, the optimal focused position is achieved. The TEM image of the tip top loaded with quantum dots has been shown in Fig. 5(a). A clear scanning TEM (STEM) image of one single quantum dot is acquired at an accelerating voltage of 300 kV [Fig. 5(b)]. The crystal structure is found to be hexagonal wurtzite along the [0001] zone axis. At the same time, the pulse laser beam (wavelength centered at 800 nm) is focused onto the quantum dots to excite two-photon fluorescence. Significant two-photon PL signals (wavelength centered at 630 nm) from the quantum dot ensemble are obtained [Fig. 5(c)], indicating the high efficiency of optical excitation and collection. The collected signals are then sent to the SPAD of the TCSPC system. The PL lifetime can be obtained, and Fig. 5(d) shows a typical lifetime of 4.3 ns for CdSe/ZnS quantum dots. Our results provide an example for *in situ* optical spectroscopy with high temporal resolution in AC-TEM, which gives structural information and PL lifetime of quantum dots simultaneously.

B. Discussions

It is always difficult to introduce focused laser beams into the commercial AC-TEM specimen chamber without modification of its components. In order to realize higher excitation efficiency and temporal resolution, the pulse beam with a smaller focal spot size and narrower pulse width should be introduced into TEM. As for our apparatus, there is no need to modify the main body of the AC-TEM for the achievement of optical measurement with high temporal resolution. Together with a 4*f* de-magnified system and the dispersion compensation module, our designed specimen holder can successfully introduce a focal spot with a size less than 2 μm and a pulse width of about 300 fs (@ 3 mW) into AC-TEM. Moreover, the holder could also be used to guide the continuous wave laser into TEM, and the dispersion compensation module is not needed under this circumstance. Importantly, to realize high excitation efficiency, the cores of the fiber bundle should be single-mode or few-mode at the working wavelength.

In this work, we conduct time-correlated single photon counting measurement for quantum dots and obtain a PL lifetime of 4.3 ns. The pulse width of the introduced laser beam is compressed into hundreds of femtoseconds, which could guarantee the fs-scale temporal resolution for time-resolved optical measurement. Further ultrafast optical spectroscopy can also be conducted such as one-color pump-probe measurement. As for two-color pump-probe measurement, a fiber bundle suitable for the two working wavelengths is needed. The time delay between the pump

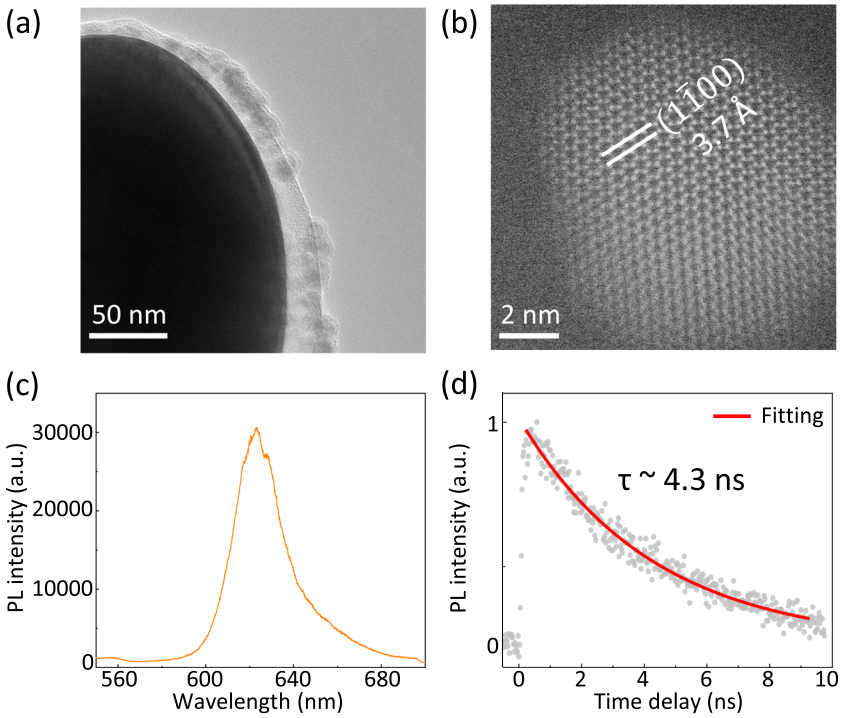

FIG. 5. *In situ* time-resolved PL of CdSe/ZnS quantum dots in AC-TEM: (a) the TEM image of the tip top loaded with CdSe/ZnS quantum dots, (b) the STEM image of the CdSe/ZnS quantum dot observed in AC-TEM, (c) the *in situ* two-photon PL spectrum of CdSe/ZnS quantum dots, and (d) *in situ* measurement of PL lifetime of CdSe/ZnS quantum dots by using TCSPC.

and the probe beam can be adjusted using translational stages and mirrors before coupling into the fiber. After collecting the probe signals, ultrafast dynamics can be revealed *in situ* in AC-TEM. Moreover, since the specimen can be affected by the electron beam to some extent, optical response from the specimen, such as PL, would be expected to change accordingly. Our apparatus can further study the influence of the electron beam to the specimen *in situ* through optical measurement and ultrafast optical spectroscopy.

V. SUMMARY

In conclusion, we have developed a specially designed specimen holder to realize *in situ* optical measurement with high temporal resolution in AC-TEM. With a fiber bundle and 4*f* de-magnified system used in the specimen holder, the focused pulse beam with a size less than 2 μm can be guided into TEM. The generated focal spot can be scanned by using translational stages, and the scanning area is larger than 75 × 75 μm^2, with a resolution of 10 nm. To compensate the positive GVD caused by glass fiber, a dispersion compensation module is used to compress the pulse width into about 300 fs at an output power of 3 mW. *In situ* TCSPC measurement of PL lifetime together with high-resolution characterization of the atomic structure of CdSe/ZnS quantum dots is conducted successfully. The achievement of focused pulse beams with a hundred-femtosecond-scale pulse width in AC-TEM enables us to conduct ultrafast optical spectroscopy. Further, one-color or two-color pump-probe measurement is promising by utilizing our apparatus, which will unveil ultrafast dynamics of nano-materials with atomic-scale spatial resolution. Our apparatus provides a practical solution for *in situ* optical spectroscopy with high temporal resolution in AC-TEM.

AUTHORS' CONTRIBUTIONS

C.L. and C.M. contributed equally to this work.

ACKNOWLEDGMENTS

This work was supported by the National Equipment Program of China (Grant No. ZDYZ2015-1), the Beijing Natural Science Foundation (Grant No. JQ19004), Beijing Excellent Talents Training Support—Top Young Individual Project (No. 2017000026833ZK11), the National Key R&D Program of China (Grant Nos. 2016YFA0300903 and 2016YFA0300804), the National Science Fund for Distinguished Young Scholars (Grant No. 52025023), the National Natural Science Foundation of China (Grant Nos. 51991340 and 51991342), the Beijing Municipal Science and Technology Commission (Grant No. Z191100007219005), the Beijing Graphene Innovation Program (Grant No. Z181100004818003), the Strategic Priority Research Program of CAS (Grant Nos. XDB28000000 and XDB33000000), and the Innovation Center of Fadiation Application, China Institute of Atomic Energy (Grant No. KFZC2018040203).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

REFERENCES

[1]E. Ruska, "The development of the electron-microscope and of electron-microscopy," Rev. Mod. Phys. **59**, 627–638 (1987).
[2]Z. L. Wang, "New developments in transmission electron microscopy for nanotechnology," Adv. Mater. **15**, 1497–1514 (2003).
[3]D. B. Williams and C. B. Carter, *Transmission Electron Microscopy: A Textbook for Materials Science* (Springer US, 2009).
[4]R. F. Egerton, *Electron Energy-Loss Spectroscopy in the Electron Microscope* (Springer US, 2011).
[5]L. J. Allen, S. D. Findlay, A. R. Lupini, M. P. Oxley, and S. J. Pennycook, "Atomic-resolution electron energy loss spectroscopy imaging in aberration corrected scanning transmission electron microscopy," Phys. Rev. Lett. **91**, 105503 (2003).
[6]P. E. Batson, N. Dellby, and O. L. Krivanek, "Sub-ångstrom resolution using aberration corrected electron optics," Nature **418**, 617–620 (2002).
[7]A. Y. Borisevich, A. R. Lupini, and S. J. Pennycook, "Depth sectioning with the aberration-corrected scanning transmission electron microscope," Proc. Natl. Acad. Sci. U. S. A. **103**, 3044–3048 (2006).
[8]N. Dellby, O. L. Krivanek, P. D. Nellist, P. E. Batson, and A. R. Lupini, "Progress in aberration-corrected scanning transmission electron microscopy," J. Electron Microsc. **50**, 177–185 (2001).
[9]M. Haider, H. Rose, S. Uhlemann, E. Schwan, B. Kabius, and K. Urban, "A spherical-aberration-corrected 200 kV transmission electron microscope," Ultramicroscopy **75**, 53–60 (1998).
[10]M. Haider, S. Uhlemann, and J. Zach, "Upper limits for the residual aberrations of a high-resolution aberration-corrected STEM," Ultramicroscopy **81**, 163–175 (2000).
[11]C.-L. Jia, M. Lentzen, and K. Urban, "High-resolution transmission electron microscopy using negative spherical aberration," Microsc. Microanal. **10**, 174–184 (2004).
[12]M. Lentzen, B. Jahnen, C. L. Jia, A. Thust, K. Tillmann, and K. Urban, "High-resolution imaging with an aberration-corrected transmission electron microscope," Ultramicroscopy **92**, 233–242 (2002).
[13]S. J. Pennycook, B. Rafferty, and P. D. Nellist, "Z-contrast imaging in an aberration-corrected scanning transmission electron microscope," Microsc. Microanal. **6**, 343–352 (2000).
[14]D. J. Smith, "Development of aberration-corrected electron microscopy," Microsc. Microanal. **14**, 2–15 (2008).
[15]S. Uhlemann and M. Haider, "Residual wave aberrations in the first spherical aberration corrected transmission electron microscope," Ultramicroscopy **72**, 109–119 (1998).
[16]K. W. Urban, "Studying atomic structures by aberration-corrected transmission electron microscopy," Science **321**, 506–510 (2008).
[17]M. Varela, A. R. Lupini, K. van Benthem, A. Y. Borisevich, M. F. Chisholm, N. Shibata, E. Abe, and S. J. Pennycook, "Materials characterization in the aberration-corrected scanning transmission electron microscope," Annu. Rev. Mater. Res. **35**, 539–569 (2005).
[18]P. W. Hawkes and J. C. H. Spence, *Science of Microscopy* (Springer, New York, 2008).
[19]M. L. Taheri, E. A. Stach, I. Arslan, P. A. Crozier, B. C. Kabius, T. LaGrange, A. M. Minor, S. Takeda, M. Tanase, J. B. Wagner, and R. Sharma, "Current status and future directions for in situ transmission electron microscopy," Ultramicroscopy **170**, 86–95 (2016).
[20]P. Chen, X. Zhong, J. A. Zorn, M. Li, Y. Sun, A. Y. Abid, C. Ren, Y. Li, X. Li, X. Ma, J. Wang, K. Liu, Z. Xu, C. Tan, L. Chen, P. Gao, and X. Bai, "Atomic imaging of mechanically induced topological transition of ferroelectric vortices," Nat. Commun. **11**, 1840 (2020).
[21]X. Li, C. Tan, C. Liu, P. Gao, Y. Sun, P. Chen, M. Li, L. Liao, R. Zhu, J. Wang, Y. Zhao, L. Wang, Z. Xu, K. Liu, X. Zhong, J. Wang, and X. Bai, "Atomic-scale observations of electrical and mechanical manipulation of topological polar flux closure," Proc. Natl. Acad. Sci. U. S. A. **117**, 18954–18961 (2020).
[22]L. Zhu, S. Chen, H. Zhang, J. Zhang, Y. Sun, X. Li, Z. Xu, L. Wang, J. Sun, P. Gao, W. Wang, and X. Bai, "Strain-inhibited electromigration of oxygen vacancies in $LaCoO_3$," ACS Appl. Mater. Interfaces **11**, 36800–36806 (2019).

[23]T. Ohmura, A. M. Minor, E. A. Stach, and J. W. Morris, "Dislocation-grain boundary interactions in martensitic steel observed through in situ nanoindentation in a transmission electron microscope," J. Mater. Res. **19**, 3626–3632 (2004).
[24]J. Sun, L. He, Y.-C. Lo, T. Xu, H. Bi, L. Sun, Z. Zhang, S. X. Mao, and J. Li, "Liquid-like pseudoelasticity of sub-10-nm crystalline silver particles," Nat. Mater. **13**, 1007–1012 (2014).
[25]Q. Yu, M. Legros, and A. M. Minor, "*In situ* TEM nanomechanics," MRS Bull. **40**, 62–68 (2015).
[26]P. Gao, Z. Kang, W. Fu, W. Wang, X. Bai, and E. Wang, "Electrically driven redox process in cerium oxides," J. Am. Chem. Soc. **132**, 4197–4201 (2010).
[27]J. Y. Huang, L. Zhong, C. M. Wang, J. P. Sullivan, W. Xu, L. Q. Zhang, S. X. Mao, N. S. Hudak, X. H. Liu, A. Subramanian, H. Fan, L. Qi, A. Kushima, and J. Li, "In situ observation of the electrochemical lithiation of a single SnO_2 nanowire electrode," Science **330**, 1515–1520 (2010).
[28]L. Wang, Z. Xu, W. Wang, and X. Bai, "Atomic mechanism of dynamic electrochemical lithiation processes of MoS_2 nanosheets," J. Am. Chem. Soc. **136**, 6693–6697 (2014).
[29]M. Sun, J. Wei, Z. Xu, Q. Huang, Y. Zhao, W. Wang, and X. Bai, "Electrochemical solid-state amorphization in the immiscible Cu-Li system," Sci. Bull. **63**, 1208–1214 (2018).
[30]J. Wang, M. Sun, Y. Liu, J. Lin, L. Wang, Z. Xu, W. Wang, Z. Yuan, J. Liu, and X. Bai, "Unraveling nanoscale electrochemical dynamics of graphite fluoride by *in situ* electron microscopy: Key difference between lithiation and sodiation," J. Mater. Chem. A **8**, 6105–6111 (2020).
[31]S. K. Eswaramoorthy, J. M. Howe, and G. Muralidharan, "In situ determination of the nanoscale chemistry and behavior of solid-liquid systems," Science **318**, 1437–1440 (2007).
[32]S. H. Oh, Y. Kauffmann, C. Scheu, W. D. Kaplan, and M. Ruhle, "Ordered liquid aluminum at the interface with sapphire," Science **310**, 661–663 (2005).
[33]K. Suzuki, M. Ichihara, S. Takeuchi, K. Nakagawa, K. Maeda, and H. Iwanaga, "*In situ* TEM observation of dislocation-motion in II–VI compounds," Philos. Mag. A **49**, 451–461 (1984).
[34]Y. Ohno and S. Takeda, "A new apparatus for *in-situ* photoluminescence spectroscopy transmission electron-microscope," Rev. Sci. Instrum. **66**, 4866–4869 (1995).
[35]Y. Ohno, T. Taishi, and I. Yonenaga, "*In situ* analysis of optoelectronic properties of dislocations in ZnO in TEM observations," Phys. Status Solidi A **206**, 1904–1911 (2009).
[36]Y. Ohno, "Development of an apparatus for *in-situ* near-field photoexcitation in a transmission electron microscope," Appl. Phys. Express **5**, 125204 (2012).
[37]M. Picher, S. Mazzucco, S. Blankenship, and R. Sharma, "Vibrational and optical spectroscopies integrated with environmental transmission electron microscopy," Ultramicroscopy **150**, 10–15 (2015).
[38]F. I. Allen, E. Kim, N. C. Andresen, C. P. Grigoropoulos, and A. M. Minor, "*In situ* TEM Raman spectroscopy and laser-based materials modification," Ultramicroscopy **178**, 33–37 (2017).
[39]H. Dong, F. Xu, Z. Sun, X. Wu, Q. Zhang, Y. Zhai, X. D. Tan, L. He, T. Xu, Z. Zhang, X. Duan, and L. Sun, "In situ interface engineering for probing the limit of quantum dot photovoltaic devices," Nat. Nanotechnol. **14**, 950–956 (2019).
[40]J. F. S. Fernando, C. Zhang, K. L. Firestein, and D. Golberg, "Optical and optoelectronic property analysis of nanomaterials inside transmission electron microscope," Small **13**, 1701564 (2017).
[41]M. Gajdardziska-Josifovska, V. Lazarov, J. Reynolds, and V. V. Yakovlev, "Wavelength dependence of laser-induced phase transformations in semiconductor quantum dots," Appl. Phys. Lett. **78**, 3298–3300 (2001).
[42]P. Gao, Z. Z. Wang, K. H. Liu, Z. Xu, W. L. Wang, X. D. Bai, and E. G. Wang, "Photoconducting response on bending of individual ZnO nanowires," J. Mater. Chem. **19**, 1002–1005 (2009).
[43]B. Xiang, D. J. Hwang, J. B. In, S.-G. Ryu, J.-H. Yoo, O. Dubon, A. M. Minor, and C. P. Grigoropoulos, "*In situ* TEM near-field optical probing of nanoscale silicon crystallization," Nano Lett. **12**, 2524–2529 (2012).
[44]V. V. Yakovlev, V. Lazarov, J. Reynolds, and M. Gajdardziska-Josifovska, "Laser-induced phase transformations in semiconductor quantum dots," Appl. Phys. Lett. **76**, 2050–2052 (2000).
[45]F. Cavalca, A. B. Laursen, B. E. Kardynal, R. E. Dunin-Borkowski, S. Dahl, J. B. Wagner, and T. W. Hansen, "*In situ* transmission electron microscopy of light-induced photocatalytic reactions," Nanotechnology **23**, 075705 (2012).
[46]B. K. Miller and P. A. Crozier, "System for *in situ* UV-visible illumination of environmental transmission electron microscopy samples," Microsc. Microanal. **19**, 461–469 (2013).
[47]J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer, Berlin Heidelberg, 2013).
[48]N. B. Abraham, F. T. Arecchi, A. Mooradian, and A. Sona, *Physics of New Laser Sources* (Springer US, 2013).
[49]W. Göbel, J. N. D. Kerr, A. Nimmerjahn, and F. Helmchen, "Miniaturized two-photon microscope based on a flexible coherent fiber bundle and a gradient-index lens objective," Opt. Lett. **29**, 2521–2523 (2004).
[50]F. Helmchen, D. W. Tank, and W. Denk, "Enhanced two-photon excitation through optical fiber by single-mode propagation in a large core," Appl. Opt. **41**, 2930–2934 (2002).
[51]H. Nakatsuka and D. Grischkowsky, "Recompression of optical pulses broadened by passage through optical fibers," Opt. Lett. **6**, 13–15 (1981).
[52]H. Nakatsuka, D. Grischkowsky, and A. C. Balant, "Non-linear picosecond-pulse propagation through optical fibers with positive group-velocity dispersion," Phys. Rev. Lett. **47**, 910–913 (1981).
[53]B. Nikolaus and D. Grischkowsky, "12× pulse-compression using optical fibers," Appl. Phys. Lett. **42**, 1–2 (1983).
[54]H. G. Winful, "Pulse-compression in optical fiber filters," Appl. Phys. Lett. **46**, 527–529 (1985).
[55]C. V. Shank, R. L. Fork, R. Yen, R. H. Stolen, and W. J. Tomlinson, "Compression of femtosecond optical pulses," Appl. Phys. Lett. **40**, 761–763 (1982).
[56]E. Treacy, "Optical pulse compression with diffraction gratings," IEEE J. Quantum Electron. **5**, 454–458 (1969).