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Probing Nanoscale Fundamental Interactions of Electrons and Quasiparticles from Optical to Terahertz Frequencies

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14. ABSTRACT
The goal of the work was to probe fundamental nanoscale properties of electrons and quasiparticles in layered materials, correlated oxides and nitrides as well as nanoscale interactions with phase change materials by performing nanospectroscopicimaging from optical to terahertz frequencies. We have demonstrated a reconfigurable hyperbolic metasurface comprising of a heterostructure of isotopically enriched hexagonal boron nitride (hBN) in direct contact with the phase-change material vanadium dioxide (VO2). We implement a novel nanoscopy tools integrated with state-of-the-art infrared lasers and Vis/IR light sources. We have introduced hyperspectral terahertz (THz) nano-imaging by combining scattering-type scanning near-field optical microscope (s-SNOM) with THz time-domain spectroscopy (TDS). We describe the technical implementations that enabled this achievement and demonstrate its performance with a heterogeneously doped semiconductor samples. We recorded for the first time THz hyperspectral image and with a spatial nanoscale spatial resolution by measuring at each pixel with a time domain spectrum covering the range from 0.4 to 1.8 THz. Fitting the spectra with a Drude model allows for measuringnoninvasively and without the need for Ohmic contactsthe local mobile carrier
concentration of the differently doped semiconductor areas. Phase separations in ternary group III-Nitride semiconductor alloys are a significant challenge that limits the efficiency and performance of optoelectronic devices. We have shown that the growth of indium-rich ternary In1xGaxN (InGaN) alloys by MOCVD exhibits strong nanoscale phase separations, reflected in compositional and gap energy fluctuations. Instability of black phosphorus (BP) surface due to chemical degradation in
ambient conditions remains a major impediment to its prospective applications.

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Abstract
The goal of the work was to probe fundamental nanoscale properties of electrons and quasiparticles in layered materials, correlated oxides and nitrides as well as nanoscale interactions with phase change materials by performing nano-spectroscopic imaging from optical to terahertz frequencies. We have demonstrated a reconfigurable hyperbolic metasurface comprising of a heterostructure of isotopically enriched hexagonal boron nitride (hBN) in direct contact with the phase-change material vanadium dioxide (VO_2). We implement a novel nanoscopy tools integrated with state-of-the-art infrared lasers and Vis/IR light sources. We have introduced hyperspectral terahertz (THz) nano-imaging by combining scattering-type scanning near-field optical microscope (s-SNOM) with THz time-domain spectroscopy (TDS). We describe the technical implementations that enabled this achievement and demonstrate its performance with a heterogeneously doped semiconductor samples. We recorded for the first time THz hyperspectral image and with a spatial nanoscale spatial resolution by measuring at each pixel with a time domain spectrum covering the range from 0.4 to 1.8 THz. Fitting the spectra with a Drude model allows for measuring—noninvasively and without the need for Ohmic contacts—the local mobile carrier concentration of the differently doped semiconductor areas. Phase separations in ternary group III-Nitride semiconductor alloys are a significant challenge that limits the efficiency and performance of optoelectronic devices. We have shown that the growth of indium-rich ternary $In_{1-x}Ga_xN$ (InGaN) alloys by MOCVD exhibits strong nanoscale phase separations, reflected in compositional and gap energy fluctuations. Instability of black phosphorus (BP) surface due to chemical degradation in ambient conditions remains a major impediment to its prospective applications. We explored robust strategies to mitigate degradation issues, and explored novel surface near-field properties of BP.

Book Chapters and Invited Reviews

1. **Y. Abate***, D. Akinwande, H. Wang, M. Snure, and S. Cronin, "Recent Progress on Stability and Passivation of Black Phosphorus," *Adv. Mat. 1704749* (2018). DOI: 10.1002/adma.201704749

2. **Y. Abate***, V.E. Babicheva, V.S. Yakovlev, and N. Dietz, "Towards Understanding and Control of Nanoscale Phase Segregation in InGaN Alloys," (Invited Book Chapter), Imperial College Press. III-Nitride Materials, Devices and Nano-Structures: pp. 183-207
DOI: 10.1142/9781786343192_0006

Refereed Journal Articles

1. M. Kotiuga, Z. Zhang, J. Li, F. Rodolakis, H. Zhou, R. Sutarto, F. He, Q. Wang, Y. Sun, Y. Wang, N. A. Aghamiri, S. B. Hancock, L. P. Rokhinson, D. P. Landau, **Y. Abate***, J. W. Freeland, R. Comin, S. Ramanathan, and K. M. Rabe, ",Carrier Localization in Perovskite Nickelates From Oxygen Vacancies" *PNAS* 201910490 (2019). https://www.pnas.org/content/116/44/21992

2. Refractive Index-Based Control of Hyperbolic Phonon-Polariton Propagation A. Fali, S. T. White, T. G. Folland, M. He, N. A. Aghamiri, S. Liu, J. H. Edgar, J. D. Caldwell ,R. F. Haglund, **Y. Abate*** *Nano Letters* 2019 19 (11), 7725-7734 DOI: 10.1021/acs.nanolett.9b02651

3. N. A. Aghamiri, F. Huth, A. J.Huber, A. Fali, R. Hillenbrand, **Y. Abate*** Hyperspectral Time-Domain Terahertz Nanoimaging," *Optics Express* 27, 24231-24242 (2019)

DOI: 10.1364/OE.27.024231

4. Y. Sun, D. Lim, M. Kotiuga, N. Badri, C. Mathew, Z. Zhang, Y. Dong, R. Kou, C. Sun, K. Ramadoss, Q. Lu , G. Vardar, W. Bowman, I. Waluyo, A. Hunt, H. Tanaka, A. N. Hattori, S. Gamage, **Y. Abate***, V. G. Pol, H. Zhou, S. Subramanian, B. Yildiz, K. M. Rabe, S. Ramanathan, "Strongly correlated perovskite lithium-ion shuttles," *PNAS* 201805029 (2018). https://doi.org/10.1073/pnas.1805029115

5. T. G. Folland, A. F., S. T. White, J. R. Matson, S. Liu, N. A. Aghamiri, J. H. Edgar, R. F. Haglund Jr., **Y. Abate***, J. D. Caldwell, "Reconfigurable infrared hyperbolic metasurfaces using phase change materials," *Nature Communications* **2018,** 9. DOI: https://doi.org/10.1038/s41467-018-06858-y

6. V. E. Babicheva, S. Gamage, L. Zhen, S. Cronin, V. Yakovlev and **Y. Abate***, "Near-field Surface Waves in Few-Layer MoS_2," *ACS Photonics 5, 2106*(2018). DOI: 10.1021/acsphotonics.7b01563

7. V. E. Babicheva, S. Gamage, M. I. Stockman, and **Y. Abate***, "Near-field edge fringes at sharp material boundaries," *Optics Express* 25, 23935, (2017). DOI: 10.1364/OE.25.023935

8. C. McGahan, S. Gamage, J. Liang, B. Cross, R. E Marvel, Richard F Haglund, and **Y. Abate***, "Geometric constraints on phase coexistence in vanadium dioxide single crystals," *IOP Nanotechnology* 28, 085701, (9pp), (2017). DOI:10.1088/1361-6528/aa5652

9. S. Gamage, A. F., N. Aghamiri, L. Yang, P.D. Ye and **Y. Abate***, Reliable Passivation of Black Phosphorus by Thin Hybrid Coating. *Nanotechnology, Volume 28, Number 26* **2017,** *28* (26).

10. **Y. Abate***, D. Seidlitz, A. Fali, S. Gamage, V. Babicheva, V. S. Yakovlev, M. I. Stockman, R. Collazo, D. Alden, N. Dietz, "Nanoscopy of Phase Separation in InxGa1-xN Alloys", *ACS Appl. Mater. Interfaces*, 2016, 8 (35), pp 23160–23166. DOI: 10.1021/acsami.6b06766

11. S. Gamage, Z. Li, V. S. Yakovlev, C. Lewis, H. Wang, S. B. Cronin, and **Y. Abate***, "Nanoscopy of Black Phosphorus Degradation", *Advanced Materials Int.* Volume: 3 Issue: 12, Article Number: 1600121. DOI: 10.1002/admi.201600121 (Cover Paper)

12. **Y. Abate***, S. Gamage, Z. Li, V. Babicheva, M. H. Javani, H. Wang, S.B. Cronin and M.I. Stockman*, "Nanoscopy Reveals surface-Metallic Black Phosphorus", *Nature Light: Science & Applications* (2016) 5, e16162. DOI: 10.1038/lsa.2016.162

13. M. K. I Senevirathna, D. Seidlitz, A. Fali, B. Cross, **Y. Abate***, N. Dietz, "Effect of AlN buffer layers on the structural and optoelectronic properties of InN/AlN/Sapphire heterostructures grown by MEPA-MOCVD" *Proc Spie* **2016,** *9954*. DOI: https://doi.org/10.1117/12.2237957

14. **Y. Abate***, R. E. Marvel, J. I. Ziegler, S. Gamage, M. H. Javani, M. I. Stockman, R. F. Haglund, "Control of plasmonic nanoantennas by reversible metal-insulator transition", *Nature Sci. Rep.* 5, 13997. (2015). DOI: 10.1038/srep13997

15. D. Seidlitz, M. K. I. S., **Y. Abate***, A. Hoffmann and N. Dietz, Optoelectronic and structural properties of InGaN grown by Migration-Enhanced, Plasma-Assisted MOCVD. *SPIE Conf. Proc. 9571, paper 9571-25, 14th Int. Conf. SSL & LED-based Illum. Systems* **2015**. DOI: 10.1117/12.2188612

Patents

None

Graduate (PhD, MS), Undergraduate and Postdoctoral Students Advised, Supported by AFOSR Grant

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Durai Murugan	Postdoc	2016-2017	Near-field microscopy	Research Associate, Raman Institute, Bengaluru - 560 080 INDIA.
Collin Lewis	Undergraduate (BSc)	2017	Sample preparation	PhD program
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Darin Mihalik	Graduate (PhD)	Fall 2018, Spring 2019	Scanning Probe Microscopy	University of Georgia
Brendan Cross	Graduate (MS)	2017	Nitrides nanoscopy	PhD Program, Georgia State Uni.
Andrea Hill	Undergraduate (BSc)	current	Sample preparation	University of Georgia
Steven Hancock	Graduate (PhD)	Current	Correlated materials	University of Georgia
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Alireza Fali	Graduate (PhD)	Current	Nano-imaging, spectroscopy	University of Georgia