Oxide Materials for Emerging Applications in Photonics: introduction to the special issue

N. Kinsey,1 R. Grange,2 B. Mendez,3 K. Sun,4 and O. L. Muskens4,\*

1Department of Electrical and Computer Engineering, Virginia Commonwealth University, Richmond, VA 23284, USA

2ETH Zurich, Department of Physics, Institute for Quantum Electronics, Optical Nanomaterial Group, Auguste-Piccard-Hof 1, 8093 Zurich, Switzerland

3Department of Physics of Materials, Faculty of Physical Sciences, University Complutense of Madrid, 28040 Madrid, Spain

3Physics and Astronomy, Faculty of Engineering and Physical Sciences, University of Southampton, SO17 1BJ, Southampton

\*o.muskens@soton.ac.uk

**Abstract:** This is an introduction to the feature issue of *Optical Materials Express* on *Oxide Materials for Emerging Applications in Photonics*.

© 2021 Optica Publishing Group under the terms of the [Optica Publishing Group Open Access Publishing Agreement](https://doi.org/10.1364/OA_License_v2)

Optics is a discipline that is inherently intertwined with materials, and many of the key breakthroughs achieved in the field have been directly enabled by material platform developments and implementations. In this vein, few material families have had a larger impact upon optics than the development of oxides. From early studies of refraction in glass lenses and magneto-optical crystals  [1] to more recent wide bandgap semiconductor light sources  [2] and metamaterial lenses  [3], oxides have literally enabled photonics; perhaps most well evidenced through the role of optical glasses and the International Year of Glass celebration in 2022  [4,5]. Despite this exceptional history of development and impact, there is still much work to be done in the development and utilization of both new and well-established oxide materials.

This feature issue intends to offer a look into some of the latest thoughts on the use of oxide materials in optical applications, generally grouped across five thematic areas: high damage threshold oxides, epsilon-near-zero (ENZ) effects, nanostructured oxides, post fabrication tuning & switching of conducting oxides, and oxides in integrated devices.

Through the rise of many new technologies in medical, defense, manufacturing, and scientific communities, high damage threshold devices are becoming increasingly important. For medical and industrial applications, damage to the active materials remains an important issue. Y. Wang et al. presents the first experimental and theoretical demonstration of the laser damage mechanism in La3Ga5.5Nb0.5O14 (LGN) crystals  [6]. They optimized the annealing condition to improve the damage threshold significantly. Additionally, a strategy to increase the laser damage fluence threshold of sputtered hafnia thin films, a key low-loss and ‘high-index’ oxide in the visible, is presented in the paper by P. B. Mirkarimi et al. [7]. The approach consists of substituting the conventional argon working gas for xenon during ion beam sputtering. As a result, a significant increase in the damage threshold is reported (1 J/cm2). This outcome would have significant implications for the advancement of ultraviolet laser systems, with potential benefits for various research fields, particularly for the enhancement of laser based inertial confinement fusion research devices.

In recent years, the rise of epsilon-near-zero (ENZ) materials  [8], such as layered metal dielectric stacks or heavily doped oxides, have been shown to provide unique light matter interactions by making use of the slow light propagation, boundary field enhancement/localization, and small phase accumulation among others, thereby opening new potential applications across various areas of photonics  [9]. Yet, one issue that remains is the large impedance mismatch with such materials, making them difficult to couple to. In the work by L. C. Wynne et al., an approach to realize anti-reflection coatings for epsilon-near-zero materials comprising multilayer silica-silver films is highlighted  [10]. The method shown improves the transmission by 20% while decreasing reflection by 50% over a 200 nm range, aiding the ability to effectively funnel light into epsilon-near-zero materials for applications across both linear and nonlinear effects. In this view, ENZ effects in the transparent conducting oxides have recently shown promise towards achieving strong and fast nonlinear optical responses, achieving unity order index modulation on picosecond scales  [11]. Yet, rigorous characterization of such materials in multiple dimensions (pump wavelength, probe wavelength, time, polarization, etc.) has been lacking. In the work by S. Benis et al., an invited contribution, multiple techniques (beam deflection, Z-scan, cross-phase modulation) have been utilized to fill this gap and provide a reliable and robust characterization to spur future development in areas of compact nonlinear devices in all platforms  [12].

The rapid rise of nanofabrication techniques in the 21st century has been a catalyst for miniaturizing/optimizing photonic devices, developing new methods to control light matter interaction, and manipulating chemical reactions, among others. Among them, modern two-photon lithography (TPL) technologies have been an effective method for fabricating 3D printing featured structures  [13]. Yet, the photoresin refractive index is mostly limited to values between 1.45-1.55, limiting potential applications and efficiencies. Through synthesizing and functionalizing titanium oxide nanoparticles into commercial resins, R. S. Ketchum et al. demonstrate an improved TPL photoresin with refractive index of 1.66 at 633 nm  [14]. Within the field of quantum technology, in particular for the generation of entangled photon sources, applications can profit from the miniaturization of metal-oxides, such as lithium niobate. Besides the thin film technology of lithium niobate on insulator that is maturing very fast  [15], N. M. H. Duong et al. use bottom-up synthesized lithium niobate microcubes to generate photon pairs without phase matching condition with an efficiency of 20 GHz/W-m  [16]. Finally, properties of metal oxides relevant for applications such as catalysis and optoelectronics are explored by A. Vázquez-López et al., an invited contribution, in their studies of the time-resolved photoluminescence properties of Li-doped TiO2 nanoparticles over a range of temperatures down to 10 K  [17]. These studies can identify thermally activated recombination pathways related to defects and surface trapping states mediated by the presence of Li.

One of the key features of many oxide materials is the ability to both dynamically and statically tune their optical properties, opening potential in high-speed active devices as well as sensors and information storage devices. In particular, the capability of reactive laser annealing (ReLA) — a technique that combines the advantages of reactive ambient thermal annealing and laser annealing—is discussed in-depth in the paper by J. A. Hillier et al.  [18]. In particular, they address the issue of finding strategies for reducing the carrier concentration of ITO films. The authors have investigated the synthesis route and the role of the laser fluence and ambient composition and performed extensive characterization in order to fully elucidate the laser-matter interactions. As a result, the approach is revealed as an effective route to selectively tune the carrier transport properties of ITO films. Reversible switching between different oxidative states is another unique benefit of oxide materials, and is shown to offer a mechanism for non-volatile switching and memory applications in work presented by Y. Gutiérrez et al.  [19]. They show that the MoO3-to-MoO2 transition is associated with large changes in the optical response ranging from dielectric to metallic behavior in parts of the spectrum. The cycling was achieved by repeated annealing under hydrogen and air. Such materials could be used for integration with photonic waveguides to realize amplitude modulators or reflection modulators in free-space. Similarly, the ability to dynamically alter the optical properties of oxides has interesting application in the area of structural color for security markers, new forms or art, and data storage [20]. While the design of static devices has been explored in recent years, the optimization of dynamic structural color systems represents a difficult design challenge with multiple objectives. In the work by P. Dai et al., an approach utilizing generative adversarial networks overcomes this challenge, realizing dynamic pixel designs which can simultaneously achieve multiple desired states based on the thermal cycling of VO2 layers embedded in a Fabry-Pérot cavity  [21]. The structure is shown to achieve a gamut of 117% of the sRGB spectrum with a 5% color coverage variation, opening new possibilities for high density data storage systems and trustable information.

Finally, the rise of integrated photonic technologies has created new demands for material waveguiding platforms, processing techniques, and integration methodologies. Silicon oxynitride (SiON) is one platform for integrated photonics which is of interest because of its ability to reduce film stress compared to pure silicon nitride. By increasing oxidation the refractive index can be continuously tuned between 1.45 - 2.00. G. Piccoli et al. demonstrate a platform based on SiON channel waveguides which combines favorable characteristics such as high optical bandgap and strong nonlinearity with dispersion engineering for a range of applications in nonlinear conversion  [22]. Yet, the top-down fabrication of oxides devices is still a challenge to achieve the highest light confinement with the lowest losses. A new post-fabrication trimming for integrated photonic circuit is demonstrated in one of the papers of this issue by A. V. Tronev et al.  [23]. They measure an increase in the extinction ratio by 27 dB from 30 dB up to 57 dB in an integrated optical Ti:LiNbO3 Mach-Zehnder modulator.

While it is impossible to capture the entire breadth of impact that oxide materials have on the optics community, the works collected herein clearly illustrate that oxide materials continue to have an outsized impact upon the latest optical science and technology. We are excited to see how these oxide materials and applications evolve in the coming years. We hope that you enjoy this special issue and express our thanks to all the authors, reviewers, and OSA staff members for their contributions to make this issue possible.

**Funding.** Air Force Office of Scientific Research (FA9550-18-1-0151, FA9950-22-1-0383); National Science Foundation (1808928); European Research Council 714837 (Chi2-nano-oxides)

**Disclosures.** The authors declare no competing interests.

References

1. E. Hecht, *Optics*, 5th ed. (Pearson, 2017).

2. H. Morkoç and Ü. Özgür, *Zinc Oxide: Fundamentals, Materials and Device Technology* (John Wiley & Sons, 2008).

3. A. C. Overvig, S. Shrestha, S. C. Malek, M. Lu, A. Stein, C. Zheng, and N. Yu, "Dielectric metasurfaces for complete and independent control of the optical amplitude and phase," Light Sci. Appl. **8**, 92 (2019).

4. U. Nations, "Glass & Innovation - International Year of Glass 2022," https://media.un.org/en/asset/k14/k14m7j47jb.

5. J. Ballato, "Optical Fiber: Through the looking glass," Opt. Photonics News2 (2022).

6. Y. Wang, F. Liang, D. Lu, S. Wang, J. Wang, H. Yu, and H. Zhang, "Laser damage mechanism and threshold improvement of nonlinear optical La 3 Ga 5.5 Nb 0.5 O 14 crystal for a mid-infrared high-intensity laser," Opt. Mater. Express **12**, 3449 (2022).

7. P. B. Mirkarimi, C. Harthcock, S. R. Qiu, R. A. Negres, G. Guss, T. Voisin, J. A. Hammons, C. A. Colla, H. E. Mason, A. Than, D. Vipin, and M. Huang, "Improving the laser performance of ion beam sputtered dielectric thin films through the suppression of nanoscale defects by employing a xenon sputtering gas," Opt. Mater. Express **12**, 3365 (2022).

8. I. Liberal and N. Engheta, "Near-zero refractive index photonics," Nat. Photonics **11**, 149–158 (2017).

9. N. Kinsey, C. DeVault, A. Boltasseva, and V. M. V. M. Shalaev, "Near-zero-index materials for photonics," Nat. Rev. Mater. **4**, (2019).

10. L. C. Wynne, C. Zhang, U. Akpan, A. Di Falco, and S. A. Schulz, "Anti-reflection coatings for epsilon-near-zero materials," Opt. Mater. Express **12**, 4088 (2022).

11. O. Reshef, I. De Leon, M. Z. Alam, and R. W. Boyd, "Nonlinear optical effects in epsilon-near-zero media," Nat. Rev. Mater. 1 (2019).

12. S. Benis, N. Munera, S. Faryadras, E. W. Van Stryland, and D. J. Hagan, "Extremely large nondegenerate nonlinear index and phase shift in epsilon-near-zero materials [Invited]," Opt. Mater. Express **12**, 3856 (2022).

13. V. Harinarayana and Y. C. Shin, "Two-photon lithography for three-dimensional fabrication in micro/nanoscale regime: A comprehensive review," Opt. Laser Technol. **142**, 107180 (2021).

14. R. S. Ketchum, P. E. Alcaraz, and P.-A. Blanche, "Modified photoresins with tunable refractive index for 3D printed micro-optics," Opt. Mater. Express **12**, 3152 (2022).

15. A. Boes, B. Corcoran, L. Chang, J. Bowers, and A. Mitchell, "Status and Potential of Lithium Niobate on Insulator (LNOI) for Photonic Integrated Circuits," Laser Photon. Rev. **12**, 1700256 (2018).

16. N. M. Hanh Duong, G. Saerens, F. Timpu, M. T. Buscaglia, V. Buscaglia, A. Morandi, J. S. Müller, A. Maeder, F. Kaufmann, A. S. Solntsev, and R. Grange, "Spontaneous parametric down-conversion in bottom-up grown lithium niobate microcubes," Opt. Mater. Express **12**, 3696 (2022).

17. A. Vázquez-López, A. Cremades, and D. Maestre, "Temperature-dependent photoluminescence of anatase Li-doped TiO 2 nanoparticles [Invited]," Opt. Mater. Express **12**, 3090 (2022).

18. J. Hillier, P. Patsalas, D. Karfardis, W. Cranton, A. Nabok, C. Mellor, D. Koutsogeorgis, and N. Kalfagiannis, "Reactive laser annealing of indium tin oxide: implications to crystal structure, defect composition, and plasma energy," Opt. Mater. Express (2022); DOI 10.1364/OME.464918.

19. Y. Gutiérrez, G. Santos, F. Palumbo, M. Modreanu, F. Moreno, and M. Losurdo, "Reversible and non-volatile metal-to-insulator chemical transition in molybdenum oxide films," Opt. Mater. Express **12**, 3957 (2022).

20. A. Kristensen, J. K. W. Yang, S. I. Bozhevolnyi, S. Link, P. Nordlander, N. J. Halas, and N. A. Mortensen, "Plasmonic colour generation," Nat. Rev. Mater. **2**, 16088 (2017).

21. P. Dai, K. Sun, O. L. Muskens, C. H. de Groot, and R. Huang, "Inverse design of a vanadium dioxide based dynamic structural color via conditional generative adversarial networks," Opt. Mater. Express **12**, 3970 (2022).

22. G. Piccoli, M. Sanna, M. Borghi, L. Pavesi, and M. Ghulinyan, "Silicon oxynitride platform for linear and nonlinear photonics at NIR wavelengths," Opt. Mater. Express **12**, 3551 (2022).

23. A. V. Tronev, M. V. Parfenov, S. I. Bozhko, A. M. Ionov, R. N. Mozhchil, S. V. Chekmazov, P. M. Agruzov, I. V. Ilichev, and A. V. Shamrai, "Local laser oxidation of titanium film for the post-fabrication trimming of photonic integrated circuits," Opt. Mater. Express **12**, 4072 (2022).