

High harmonic generation from a degenerate plasma

Ting S. Luk^{1,7}, Yuanmu Yang², Jian Lu³, Alejandro Manjavacas⁴, Hanzhe Liu², Kyle Kelly⁵, Jon-Paul Maria⁶, Michael B. Sinclair⁷, Shambhu Ghimire³, Igal Brener^{1,7}

¹Center for Integrated Nanotechnologies, Sandia National Laboratories, Albuquerque, NM, USA

²State Key Laboratory for Precision Measurement Technology and Instruments, Department of Precision Instrument, Tsinghua University, Beijing 100084, China

³Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California, 94025, USA

⁴Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico 87131, USA

⁵Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC 27695

⁶Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802

⁷Sandia National Laboratories, Albuquerque, New Mexico 87185, USA

Email: tsluk@sandia.gov

Abstract—Plane electromagnetic waves can interact strongly with a spatially confined degenerate plasma in a thin layer of transparent conductive oxide at its epsilon-near-zero wavelength. Using high mobility CdO:In conductive oxide, up to 9th harmonic of the fundamental pump wave was observed. This approach offers great flexibility for input laser wavelengths.

Keywords—epsilon-near-zero, degenerate plasma, harmonic generation, conductive oxide.

I. INTRODUCTION

High harmonic generation has been used to generate extreme ultra-violet light sources to probe fast electron dynamics in the attosecond time scale[1]. Harmonic orders as high as thousands have been demonstrated in high pressure gas based on the mechanism that electrons produced by tunnel ionizations are driven more strongly by the external electromagnetic field than the coulomb field of its parent ion[2]. Similarly, electrons in doped semiconductors or transparent conductive oxides can be driven by laser field and may interact or scatter from the ionic fields in the crystal to create highly nonlinear electronic motions and hence producing harmonic fields efficiently. Because of high electron and ion densities in solid, and the ions are essentially locked in a lattice space, the interaction dynamics can be quite different from gaseous media. Indeed, high harmonic generation from graphene[3], ZnO[4], MoS₂[5] was observed. In this paper, we explore high doped semiconductor materials where the ionic fields are screened, and electrons are largely free. The intensity required to produce significant nonlinear response is lower. For the CdO:In, 9th harmonic was observed using intensities of 10GW/cm² rather than TW/cm².

II. RESULTS

The sample under studied is a 75nm thick In doped CdO film on MgO crystal substrate with a carrier density of $2.8 \times 10^{20} \text{ cm}^{-3}$ and an electron mobility of $200 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. The sample is coated with 200nm thick Au and the laser field is coupled to the sample from the substrate side to achieve the maximum field enhancement. This structure possesses a mode which can be excited with a p-polarized $2.08 \mu\text{m}$ femtosecond laser pulse with near zero reflectivity as shown in Figure 1.

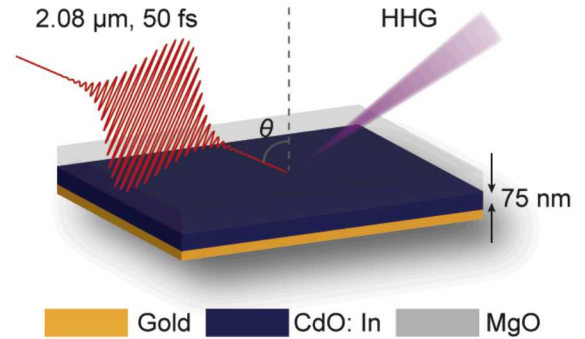


Figure 1. Schematic illustration of the excitation geometry of the sample.

The pump and generated harmonic radiations in the specular reflection direction of the pump radiation. The harmonic radiation are separated and filtered before being dispersed by a spectrometer to produce a spectrum shown in Figure 2.

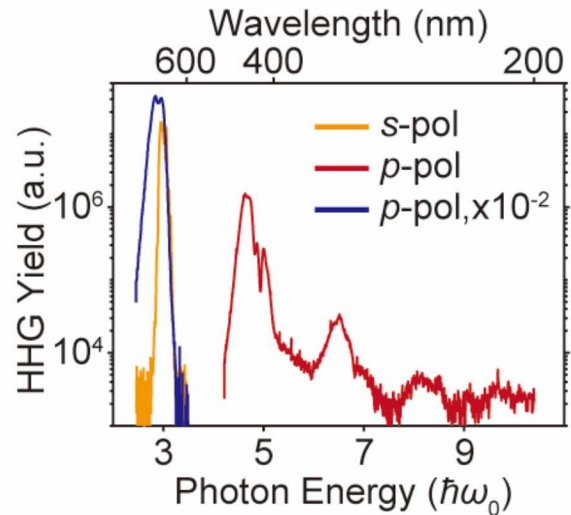


Figure 2. The harmonic spectrum from the CdO:In structure excited by $2.08 \mu\text{m}$ at an intensity of 11.3 GW/cm^2 .

Figure 2 shows the 3rd harmonic wave generated from p-polarized pump is two orders of magnitude stronger than S-polarized, indicating that the free electrons dominates the nonlinear response. Furthermore, the harmonic fields frequencies are red shifted and broadened compared to S-polarized exciting field. Using a simple two electron model[6] to account for the laser heating effects of the electrons which causes an increase in the averaged effective mass can qualitatively explain the red shift behavior. A more sophisticated dynamical model is needed for quantitative agreement.

ACKNOWLEDGMENTS

This work performed at Sandia National Laboratories was supported by the US Department of Energy (DOE), Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, and performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE's National Nuclear Security Administration under contract DE-NA-0003525. The work performed at SLAC was primarily supported by the US Department of Energy, Office of Science, Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division through the Early Career Research Program. Y.Y. acknowledges support from the Youth Thousand Talent program of China. A.M. acknowledges the National Science Foundation (Grant ECCS-1710697).

REFERENCES

- [1] F. Krausz and M. Ivanov, "Attosecond physics," *Reviews of modern physics*, vol. 81, pp. 163-234, 2009.
- [2] T. Popmintchev, M. C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Alisauskas, G. Andriukaitis, T. Balciunas, O. D. Mucke, A. Pugzlys, A. Baltuska, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernandez-Garcia, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, "Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers," *Science*, vol. 336, pp. 1287-1291, Jun 2012.
- [3] J. D. Cox, A. Marini, and F. J. G. de Abajo, "Plasmon-assisted high-harmonic generation in graphene," vol. 8, p. 14380, 2017.
- [4] 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," *Nat Phys*, vol. 7, pp. 138-141, 2011.
- [5] H. Liu, Y. Li, Y. S. You, S. Ghimire, T. F. Heinz, and D. A. Reis, "High-harmonic generation from an atomically thin semiconductor," *Nat Phys*, vol. 13, pp. 262-265, 2017.
- [6] P. Guo, R. D. Schaller, J. B. Ketterson, and R. P. H. Chang, "Ultrafast switching of tunable infrared plasmons in indium tin oxide nanorod arrays with large absolute amplitude," *Nat Photon*, vol. 10, pp. 267-273, 2016.