Usage Report for Fiscal Year 2024

Design of nonlinear optical crystals

Name:

Zhi Li (1), ○Toshiaki Iitaka (2)

Laboratory at RIKEN:

(1) RIKEN Cluster for Pioneering Research, Theoretical Quantum Physics Laboratory

(2) RIKEN Center for Computational Science, Discrete Event Simulation Research Team

1. Background and purpose of the project, relationship of the project with other projects

The terahertz (THz) electromagnetic spectrum spans frequencies from approximately 100 GHz to 30 THz, lying between the microwave and far-infrared regions. The development of THz technologies, including THz generation, detection and modulation, is driven by the need for various applications, such as communication, imaging, and spectroscopy. Generally speaking, there are three different recipes for THz detection. In the view of photoelectric effect, THz excites valence electron and the electron-hole generation can be read in form of current or voltage signal, corresponding to photoconductive and photovoltaic types of THz detectors, which just measure the amplitude of THz field and omit the phase information. Moreover, due to the low photon energy of THz radiation, which is much smaller than the typical semiconductor band gap, efficient electron-hole pair generation for photoconductive detection is not feasible. Alternative detection methods based on current response induced by Berry curvature mechanisms, such as injection current or Berry curvature dipole-induced Hall currents in topological semimetals. These detection strategies are inherently limited by the need for direct photon-electron interaction. In the view of nonlinear optic effect, difference-frequency generation can generate new signal with low frequency which is the frequency difference between incident THz signal and local reference signal. Frequency mixer is the core element of this coherent detection system which can measure both the amplitude and phase information of THz signal. Both nonlinear optic crystals or devices can play the role of frequency mixer. The utilized nonlinear optic crystal should have larger nonlinear optic coefficient within THz band for efficient difference-frequency generation. For mixer consisting of superconductor junction, cryogenic device is necessary for superconductor-insulator-superconductor junction to achieve high signal-noise ratio. Atomic vibrations, or phonon excitations, induced by temperature or THz radiation can distort the lattice potential, thereby altering the electronic transport and optical properties of materials through electron-phonon coupling (EPC). The EPC effect underlies various phenomena in solids, including conventional superconductivity, temperature-dependent resistivity in metals, carrier mobility in semiconductors, polaron formation, and phonon-assisted optical processes. Since typical phonon frequencies are in the THz range, THz radiation is particularly effective at exciting atomic vibrations. If the phonon excitation is strongly populated under THz irradiation, electronic or thermal signal can be utilized for THz detection. In principle, the temperature- or THz-induced modulation of electronic and optical properties via EPC offers a promising alternative strategy forefficient THz detection, distinct from the direct photon-electron interaction mechanism and difference-frequency generation. Conventional detectors, such as Golay cells, bolometers, and pyroelectric sensors—rely on thermal responses, necessitating cryogenic cooling and exhibiting relatively slow operational speeds. In general, we can design THz detection within the concepts of electronic excitation, difference-frequency generation, and phonon excitation.

Although lots of standalone THz detectors have been designed based on different detection principles, room-temperature integrated THz device for efficient detection remains a technological challenge. Since silicon crystal is centrosymmetric, the second order nonlinear optic effect is vanishing, and heterodyne measurement is not efficient. Additionally, the band gap of silicon is much larger than the single photon energy of THz field, electron excitation is also not feasible. In the past decade, there has been a significant surge of progress in enabling integrated, compact and efficient chip-scale THz technology. Aluminum nitride (AlN), as a material widely used in the Micro-Electro-Mechanical Systems (MEMS), has a wide bandgap of 6.2 eV (some reference is necessary), allowing operation from deep UV to mid-infrared. AlN is also compatible with complementary metal–oxide semiconductor (CMOS) fabrication processes, making it a promising material for future foundry-level PIC manufacturing. The larger band gap makes it impossible for direct electron excitation. In this work, we propose a novel strategy for room-temperature THz detection based on measuring the static electronic polarization induced by THz field driven atomic displacement.

2. Specific usage status of the system and calculation method

We perform the first-principles calculation based on density functional theory, and Wannier function on Hokusai system, including geometric optimization, electronic structure, and optical properties (including both linear and nonlinear response theory).

3. Result

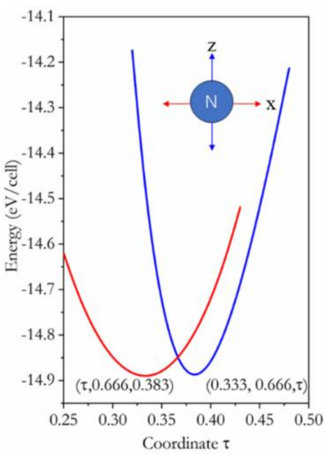

FIG. 1: Calculated total energy with off-centering atomic displacement of N in *wz* AlN with space group *P63mc*.

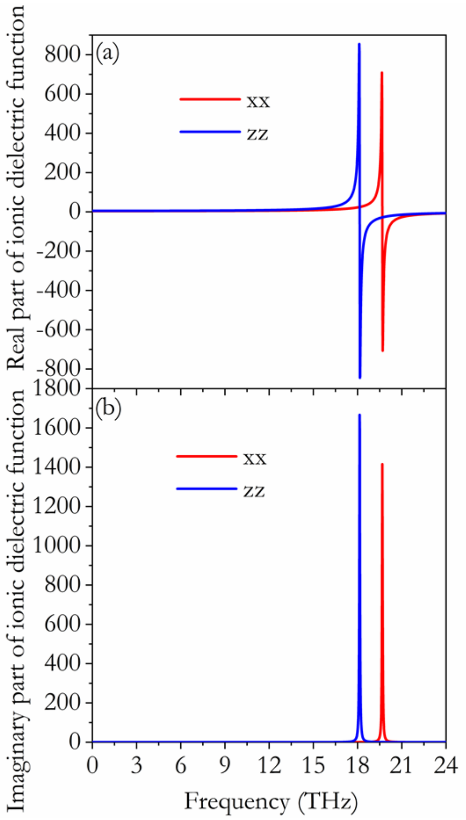

FIG. 2: Calculated real part (a) and imaginary part (b) of ionic dielectric function of *wz*-AlN with space group *P63mc*. The dielectric function contributed by ion presents two peaks of resonant absorption near f=18 THz and f=20 THz ($\omega = 2\pi f$) and weak anisotropy.

4. Conclusion

In summary, diverging from conventional approaches based on electronic excitation or frequency mixing, we propose a novel THz detection strategy that measures atomic vibration-induced static nonlinear electronic polarization on the surface of wz semiconductors. Based on model Hamiltonian analysis, we demonstrate that atomic displacement driven by a THz field at frequency ω induces static nonlinear electronic polarization through three mechanisms: nonlinear optic effect, acousto-optic effect and nonlinear phononics effect. Our numerical calculation also demonstrates that the static polarization is dominated by nonlinear acoustics effect. This work provides a novel framework for THz detection, leveraging static nonlinear polarization induced by atomic vibrations.

５. Schedule and prospect for the future

There are two important topics in future. One is developing the nonlinear optical response theory within the concept of quantum geometry tensor and computational method. Additionally, we will explore better nonlinear optic crystals with both large band gap and second order nonlinear coefficient.

６. If no job was executed, specify the reason.

Fiscal Year 2024 List of Publications Resulting from the Use of the supercomputer

[Paper accepted by a journal]

1. Zhi Li, Hussain Irshad, Yu Gu, Miriding Mutailipu, Toshiaki Iitaka, Haibin Su, and Xiangang Wan, Atomic-vibration-induced nonlinear electronic polarization for terahertz detection, Phys. Rev. B **111**, 085201 (2025).

[Conference Proceedings]

[Oral presentation]

[Poster presentation]

[Others (Book, Press release, etc.)]