Usage Report for Fiscal Year 2022

Project Title:

Prediction of Crystal Structure and Properties

Name:
Zhi Li (1), John S. Tse (2), Yanming Ma (2), ○Toshiaki Iitaka (3),

Laboratory at RIKEN:
(1) RIKEN Cluster for Pioneering Research, Theoretical Quantum Physics Laboratory
(2) RIKEN Center for Emergent Matter Science, First-Principles Materials Science Research Team
(3) RIKEN Center for Computational Science, Discrete Event Simulation Research Team

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

Electro-optic modulator (EOM), which modulates the phase, frequency, amplitude, or polarization of light by low frequency electric signal within the principle of EO effect, is one fundamental active device in integrated photonics applications, including intrachip data transmission, neuromorphic logic optical chips, and photonic integrated circuits for quantum computing. The EO effect describes the change of complex refractive index (CRI) of materials, in response to an electric field that varies slowly with time compared with the frequency of light. The real part of CRI determines the phase velocity, while the imaginary part determines the optical attenuation of light propagating in the medium. In practice, phase modulation is favored for high speed and low power optical device, i.e., we usually adopt EO crystals with relatively large band gap and relatively small attenuation coefficient at working frequency, which usually is 1550 nm for fiber-optic communication. The linear electro-optic (LEO) effect, or Pockels effect describes the change of dielectric tensor. The complex EO coefficient usually has a symmetric (antisymmetric) real (imaginary) part described the second order response under optical field with linear (circular) polarization. The real part results from the mechanism shift vector which characterizes the position difference between the Wannier centers of valance and conduction bands, while the imaginary part resulting from the curvature tensor associating with the band structure near the Fermi level. In the case of large band gap and low frequency, field strength dependent dielectric function is real and optical absorption is neglectable. For excellent EO crystals, large EO coefficient is necessary for relatively low half-wave voltage. Semiconductor $LiNbO_3$ (LN) which has relatively large band gap 3.61 eV and LEO coefficient about 32 pm/V at input wavelength 1550 nm, is the main commercial EO crystal, which is widely used for EOM device. For semiconductors with relatively small band gap, larger EO coefficients are feasible, such as the $BaTiO_3$ crystal. However, this enhancement of EO coefficient is not meaningful because of the sacrifice of band gap. It will be more significant if we can enhance both the EO coefficient and band gap simultaneously. It is challenging work to design novel EO crystals with both large band gap and EO coefficient. This work will explore excellent EO crystal with both large band gap and LEO ecoefficiency. In this work, we start from the two-band model of massive Weyl fermion, and predict high LEO coefficient resulting from the enhancement of orbital hybridization under high pressure. Then, we take the silicon nitride (Si_3N_4) electron-optical crystal as protype, and replace one Si atom by two alkaline-earth metal atoms and predict that both $SrSiN_2$ and $BaSiN_2$ under high pressure will crystalize into orthorhombic structure with the space group $Pna2_1$ of $MgSiN_2$ under ambient pressure by the first-principles calculation.

Further, we calculate the band structures and EO coefficients of isostructural $IISiN_2$ (II = Mg, Sr, Ba) to justify our proposal, viz., high pressure is an efficiently approach to enhance the LEO coefficient.

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

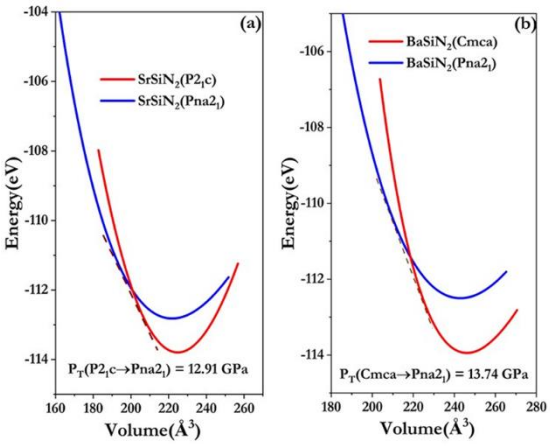

Fig. 1. The calculated EOS of SrSiN2 with space groups Pna21 and P21c (a), and EOS of BaSiN2 with space groups Pna21 and Cmca (b). Both SrSiN2 and BaSiN2 will crystalize into orthorhombic structure with space group Pna21 under high pressure, and the phase transition pressure is 12.91 and 13.74 GPa, respectively.

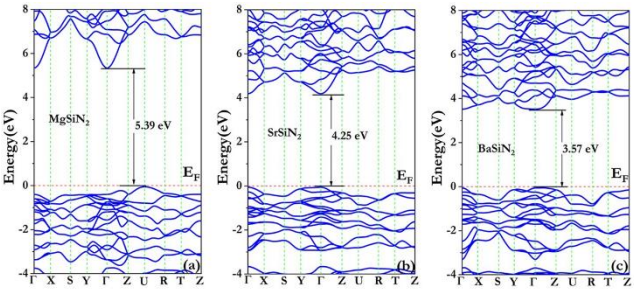

Fig. 2. Band structures of MgSiN2 (a), SrSiN2 (b) and BaSiN2 (c) with space group Pna21 by first-principles calculation with MBJ approximation.

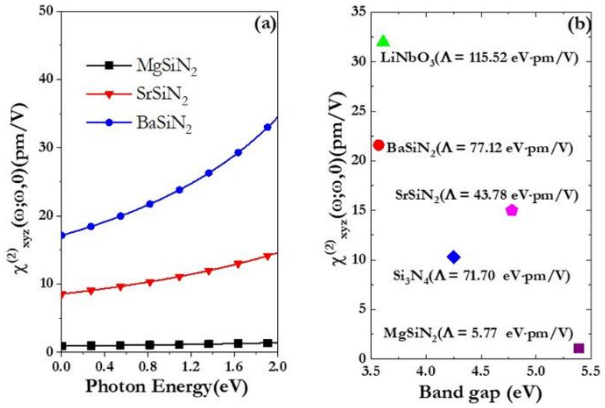

Fig. 3. (a) Calculated frequency-dependent LEO coefficient of MgSiN2, SrSiN2 and BaSiN2 with space group Pna21 contributed by shift current, where MgSiN2, SrSiN2 and BaSiN2 are represented by a black square, red triangle and blue circle. (b) The relationship between the band gap and LEO at input wavelength 1550 nm of MgSiN2, SrSiN2, BaSiN2, Si3N4 and LN.

4. Conclusion

From the theoretical analysis of nonlinear optical response within shift vector mechanism, we conclude that orbital hybridization or bond strength also affect the LEO coefficient. The first-principles calculations reveal that both SrSiN2 and BaSiN2 under high pressure condition will crystalize into orthorhombic structure with the same space group Pna21 of MgSiN2 under ambient pressure. Both SrSiN2 and BaSiN2 have remarkable large band gap about 4.25 eV and 3.57 eV, respectively, which are close to the band gap of Si3N4 and LiNbO3. The calculated second-order susceptibility of BaSiN2 (SrSiN2) at input wavelength 1550 nm is 21.60 pm/V (10.30 pm/V), which is larger that of LiNbO3 (Si3N4). Our calculated results justified our design principle, viz. high pressure can enhance the bond strength and the LEO coefficient. This work is significant for the design of excellent EO crystals with both large band gap and LEO coefficient.

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 EO crystals with both large band gap and LEO coefficient.

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

Fiscal Year 2022 List of Publications Resulting from the Use of the supercomputer

[Paper accepted by a journal]

Zhi Li, Yuewen Gao, Yu Gu, Shengli Zhang, Toshiaki Iitaka, and W. M. Liu, Berry curvature induced linear electro-optic effect in chiral topological semimetals, PHYSICAL REVIEW B 105, 125201 (2022).

[Conference Proceedings]

[Oral presentation]

[Poster presentation]

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