Nonlinear Optical Responses in α-Type Organic Salt

Keisuke Kitayama[1,2], Masao Ogata[1,3]

[1] *Department of Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan*

[2] *RIKEN Center for Emergent Matter Science, Wako, Saitama, 351-0198, Japan*

[3] *Trans-scale Quantum Science Institute, University of Tokyo, Bunkyo-ku, Tokyo 113-0033, Japan*

kitayama@hosi.phys.s.u-tokyo.ac.jp

Nonlinear optical responses, such as shift current, have been extensively explored from the perspectives of both fundamental science and electronic applications. However, nonperturbative effects in multiband systems are not well understood.

In this talk, we investigate the shift current induced by linearly polarized light in α-(BEDT-TTF)$_2$I$_3$ [see Fig. 1(a)] [1]. In our previous studies, we have theoretically predicted various photoinduced topological phase transitions in this material [2-5]. By applying the perturbation theory, we determine the dependencies of the shift current on the frequency of light. Notably, we discover that the direction of the shift current strongly depends on the frequency of light, and this unique dependence is attributed to multiband effects. Furthermore, we explore the nonperturbative effects of the shift current using the Floquet Hamiltonian [see Fig. 1(b)]. Our findings reveal a sign change in this response, a phenomenon not observable when considering only the second-order response. We discuss the limitations of both the equation derived by the perturbation theory and the one derived by Morimoto and Nagaosa [6] when the light intensity is large.

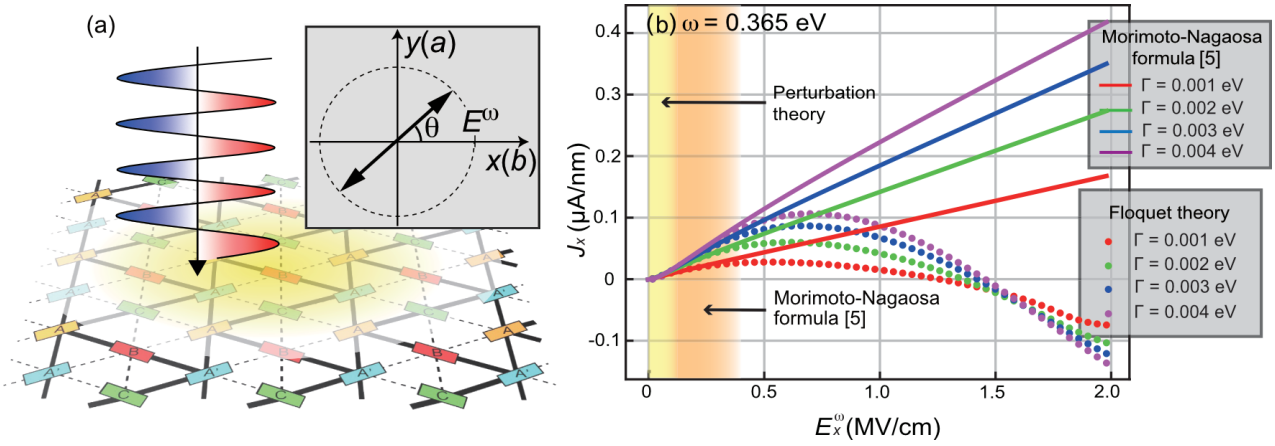

Fig.1. (a) Schematic illustration of α-(BEDT-TTF)$_2$I$_3$ under irradiation with linearly polarized light. (b) Nonlinear optical responses in photodriven α-(BEDT-TTF)$_2$I$_3$.

References

[1] K. Kitayama and M. Ogata, arXiv:2311.07176 (2023).
[2] K. Kitayama and M. Mochizuki, Phys. Rev. Research **2**, 023229 (2020).
[3] K. Kitayama, M. Mochizuki, Y. Tanaka, and M. Ogata, Phys. Rev. B **104**, 075127 (2021).
[4] K. Kitayama, Y. Tanaka, M. Ogata, and M. Mochizuki, J. Phys. Soc. Jpn. **90**, 104705 (2021).
[5] K. Kitayama, M. Ogata, M. Mochizuki, and Y. Tanaka, J. Phys. Soc. Jpn. **91**, 104704 (2021).
[6] T. Morimoto and N. Nagaosa, Science Advances **2**, e1501524 (2016).