Multiscale phenomena in molecular matter



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Ultrafast photo-control of ferroelectric order in organic and inorganic strongly correlated matters: Role of hidden state

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In spite of various attractive natures of PIPT, the research is facing a difficult and essential problem, i.e. can we realize and identify a new phase of solid based on novel lattice structure which is unique for the photo-excited condition so called as a 'hidden phase'? This 'hidden phase'with electronic and structural order realized only by optical excitation is important merit of PIPT process for achieving ultrafast and sensitive phase control via pure photonic channel free from thermal effect. We have demonstrated that 'Hidden State'really plays an essential role in PIPT process based on realistic examples. The gigantic photo-induced spectral changes in Nd_{0.5}Sr_{0.5}MnO₃ (NSMO) thin film has been attributed to the photo-induced formation of a new Charge-Orbital Ordered State based on the ps time-resolved X-ray diffraction (TR-XRD) combined with fs spectroscopic measurement [1]. We also demonstrated that such a hidden state also plays a key role in the gigantic photo-responses of organic [2,4] and inorganic [3] correlated systems. In case of $(EDO-TTF)_2PF_6$ crystals, structural changes accompanied with the photo-induced conversion of Charge Ordered patter can be probed by fs time-resolved electron diffraction and IR measurements [2]. In the present study, we report the research on the role of photo-induced 'Hidden State'in exotic ferroelectric crystals utilizing linear (UV-IR region) and nonlinear spectroscopic techniques. In the case of proton- π electron coupled organic ferroelectric system, occurrence of photo-induced electronic-proton coupled structural change has been confirmed utilizing timeresolved IR spectroscopy. In addition, SHG intensity has been modulated about 30% within 100 fs after the photo-excitation. Similar large photo-modulation in nonlinear optical property has been also confirmed in inorganic Orbital-Lattice-Spin coupled system. We discuss the importance of structural dynamics of 'Hidden State with short life-time for the development of a new class of photonic materials.

References

[1] H. Ichikawa et al., Nature Materials 10 (2011) 101.

[2] M. Gao et al., Nature 496 (2013) 343.

[3] R. Fukaya et al., Nature Commun. 6 (2015) 8519.

[4] T. Ishikawa et al., Science 350 (2015) 1501.

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