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Real-time observation of dynamic modulations over a ferro-rotational charge density wave[1]

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Dynamic control over phases of matter with electromagnetic (EM) radiation receives increasing popularity in recent years because of both its ultrafast time scale and its access to thermodynamically unapproachable phenomena. Till now, the realization of dynamic manipulation of unconventional orders, such as a high-rank multipolar order, awaits to be explored largely because the coupling between multipolar orders and EM fields is nonlinear and nontrivial. In this talk, using the commensurate charge density wave (CCDW) in 1T-TaS2 as the archetype, we demonstrate the dynamic control over the ferro-rotational order, the antisymmetric components of the second-rank electric quadrupolar order. We first confirm the ferro-rotational nature of CCDW in 1T-TaS2, broken mirrors but preserved inversion, by performing temperature-dependent rotation anisotropy-second harmonic generation (RA-SHG). We then track in real time the dynamic modulation of this ferro-rotational CCDW order, using time-resolved-RA-SHG (tr-RA-SHG) that adopts the optical-pump, RA-SHG-probe scheme. We find that this ultrafast modulation manifests itself as the breathing and the rotation of RA-SHG patterns at three different frequencies in the neighborhood of the previously reported CCDW amplitude mode frequency, with the mode of the highest (lowest) frequency primarily in the breathing (rotation) channel and the middle one in both channels. We further reveal a sudden shift of these three frequencies and a dramatic increase in the breathing and rotation magnitudes across a critical pump fluence of ˜0.5 mJ/cm2, by performing fluence dependent tr-RA-SHG.

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