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Ultrafast mid-infrared intraexcitonic spectroscopy of monolayer MoS2 SOONYOUNG CHA, School of Electrical and Electronic Engineering, Yonsei University, Seoul, Korea, JI HO SUNG, MOON-HO JO, Division of Advanced Materials Science, Pohang University of Science and Technology (POSTECH), Pohang, Korea, HYUNYONG CHOI, School of Electrical and Electronic Engineering, Yonsei University, Seoul, Korea — The optical properties of transition metal dichalcogenide (TMD) are currently active research topics for understanding the two-dimensional nature of carrier dynamics. For monolayer TMDs, reduced dielectric screening invokes strong Coulomb interactions, which lead to the large exciton binding energy. Recent theory predicts that the subset of excitons is much richer, such that internal transitions between excitons (ex. 1s, 2s, 2p) should prevail the photo-induced optical response. Here, we performed ultrafast optical pump and mid-infrared (IR) probe spectroscopy to investigate transient intraexcitonic dynamics in a monolayer MoS2. To obtain a complete excitonic dynamics, the probe photon energy is tuned over a broad range from mid-IR to IR, 0.24 eV to 0.66 eV. Our study reveals that the mid-IR responses exhibit photo-induced absorption after 3.1 eV pump excitation, which then relax within tens of picosecond and show multiple intraexcitonic and interexcitonic transitions. Our experiment shows that the fast decay component of the dynamics closely follows transient dynamics of so called A-exciton population, suggesting the photo-induced absorption indeed originates from the internal excitonic transitions.

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