Intense femtosecond photoexcitation of bulk and monolayer MoS_2

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Transition metal dichalcogenides (TMDs) are layered compounds like graphite that can be reduced from three-dimensions to two-dimensional (2D) form up to the monolayer limit due to their strong in-plane bonding and weak interlayer van der Waals coupling. In contrast with their indirect gap character in the bulk, single layer TMDs are direct gap semiconductors with great promise for optoelectronic and photonic devices. Towards the development of such devices, the investigation of 2D materials response under intense photoexcitation by ultrashort pulses, as well as of their ultrafast optical properties, is undoubtedly important.

In this work, the effect of intense femtosecond laser excitation on the structure of bulk and monolayer MoS_2 (optical gaps of 1.30 and 1.90 eV respectively), under conditions ranging from lattice heating to material damage is systematically investigated. The evolution of the Raman active A_{1g} (out of plane) and $E_{2g}{}^1$ (in plane) vibrational modes was recorded as a function of irradiation intensity and total exposure time. Experiments reveal large differences in the ultrafast laser excitation response of monolayer compared to the bulk, as far as the lattice distortion as well as the lattice morphology at the onset of optical damage. The single-pulse optical damage threshold was determined for the monolayer and bulk under 800 nm and 1030 nm pulsed laser irradiation and the role of two-photon versus one photon absorption effects is discussed.

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