Non-Linear Optical Imaging and Spectroscopic Diagnostics of 2D Transition Metal Dichalcogenides

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The interest in two-dimensional (2D) materials has been steadily increasing since the discovery of graphene, a material with fascinating properties and great potential for various applications. Transition metal dichalcogenides (TMDs) with the form MX_2 (M: W, Mo and X: S, Se, Te) exhibit a structure very similar to that of graphene and have attracted significant attention due to their extraordinary physical properties. Chlorine-doped tungsten disulfide monolayer (1L-WS_2) with tunable charge carrier concentration has been realized by pulsed laser irradiation of the atomically thin lattice in a precursor gas atmosphere. This process can sufficiently control the carrier density and consequently the PL emission of WS_2 single layers. Besides, it gives rise to a systematic shift of the neutral exciton peak towards lower energies, indicating the reduction of the crystal's electron density. The capability to progressively tune the carrier density upon variation of the exposure time is demonstrated; this implies that the Fermi level shift is directly correlated to the respective electron density modulation due to the chlorine species. Notably, this electron withdrawing process enabled the determination of the trion binding energy of the intrinsic crystal. It is found that the effect can be reversed upon continuous wave laser scanning of the monolayer in air. Density functional theory calculations indicate that chlorine physisorption is responsible for the carrier density modulation induced by the pulsed laser photochemical reaction process.

Furthermore, we use nonlinear laser-scanning optical microscopy in atomically thin TMDs to reveal, with ultra-high resolution, information about the distribution of armchair directions and their degree of organization in the 2D crystal lattice. In particular, polarization-resolved second harmonic generation (PSHG) imaging in monolayer WS_2 determines with high-precision the orientation of the main crystallographic axis (armchair). By performing a pixel-by-pixel mapping of the armchair orientations of WS_2 triangular islands on a large CVD-grown sample area, we are able to distinguish between different domains, locate their boundaries and unveil their detailed structure. To do that, we fit experimental PSHG images of sub-micron resolution into a generalized theoretical model and we acquire the armchair orientation for every pixel of the image. This allows us to measure the mean orientational average of armchair angle distributions from specific regions of interest and consequently to define the standard deviation of these distributions as a crystal quality factor.

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