Owing to their wide range of exceptional properties, two-dimensional (2D) materials have emerged as exciting media for the development of highly functional optoelectronic devices. Of these materials, the transition-metal dichalcogenides (TMDCs) are of particular interest as, unlike graphene, they are often semiconductors, and their optoelectronic properties can be tuned quite dramatically by controlling the material thickness. For example, in some materials it is possible to tune the electronic bandgap from indirect in a few layers, to direct in a single layer, of significant interest for the development of lasers. Here we focus our investigations on few-layer molybdenum disulfide (MoS2) materials for use in nonlinear optical applications. In this material, as well as controlling the bandgap, the layer thickness can be used to control the size of the nonlinear coefficients, with the second order nonlinearity turning on when the layer number is odd [1].

The MoS2 films were grown by chemical vapor deposition onto a 280 nm SiO2/Si substrate. The films were subsequently coated with a 1-μm-thick PVB layer to support the material and improve its durability. The PVB-coated MoS2 was then separated from the SiO2/Si substrate using an ultrasonic bubbling method [2]. To enhance the light-matter interactions with the ultra-thin MoS2 films, these were positioned onto specially designed low-loss side-polished optical fibers, as described in [3]. This geometry allows for the light propagating in the fiber to couple to the material over extended interaction lengths, on the order of ~1cm. Significantly, the PVB coating helps to further improve coupling to the 2D films by drawing the core guided mode into the high-index layer.

The transmission properties of the MoS2 fiber device were characterized using several different laser sources. Firstly, the linear properties were probed using a tunable CW source. Rotating the polarization revealed the expected polarization-dependent absorption, with the TE mode interacting more strongly [4]. The wavelength was then tuned to obtain the transmission spectrum shown in Fig. 1(a). The strong wavelength dependence of the interaction, with a > 30 dB extinction ratio between 1410 nm-1570 nm, suggests that there is resonant coupling between the fiber mode and the 2D material. This is in contrast with our previous measurements in graphene-based devices, which exhibited a flat absorption behavior [3]. This resonant coupling strongly enhances light-MoS2 interaction and the device’s nonlinear behavior. The second set of measurements then focused on characterizing the nonlinear properties of the film using a high-power 1540 nm fiber laser (duration of 750 fs FWHM and repetition rate of 40 MHz). Fig. 1(b) shows the normalized output power as a function of coupled input power, which clearly exhibits the onset of nonlinear absorption saturation. This result indicates the suitability of the MoS2 materials for the development of high-speed all-optical modulators, similar to what has been achieved in graphene. By monitoring the output from these measurements using an OSA, clear spectral broadening was also observed, as displayed in Fig. 1(c). This level of broadening is remarkable considering the incredibly small volume of MoS2 that the light is interacting with. We believe that the strong interaction measured in this device in this wavelength region provides clear evidence of the resonance-enhanced light-matter interaction. Thus, with further optimization of the device design and number of material layers, we predict that these structures will find use in wide-ranging nonlinear optical applications, including those based on the second order processes (e.g., second harmonic generation).

![Fig. 1](attachment:fig1.png)

(a) Absorption spectrum of the MoS2 device. (b) Measured saturable absorption curve and the corresponding fitting curve. (c) Output spectral evolution at different input average power to illustrate spectrum broadening.

References