Dynamic dielectric permittivity tensor of in-plane hyperbolic van der Waals $MoOCl_2$ and emergent chiral photonic applications

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Van der Waals (vdW) crystals offer unique opportunities for nanophotonic applications owing to their intrinsic optical anisotropy. While most of them exhibit uniaxial anisotropy, some representative families display pronounced in-plane biaxial anisotropy. Among them, vdW oxychlorides stand out due to the in-plane crystallographic axes of fundamentally different physical character—metallic and dielectric at optical frequencies. Recently, vdW $MoOCl_2$ was identified as a natural in-plane hyperbolic crystal supporting visible (Vis)–near-infrared (NIR) plasmon-polaritonic modes and exhibiting colossal optical anisotropy [1–3]. Here, we present the accurate dynamic dielectric permittivity tensor components of vdW $MoOCl_2$ in ultraviolet (UV) to Vis spectral region, partially extending into NIR. In particular, we employ imaging spectroscopic micro-ellipsometry to extract the complex dielectric permittivity tensor using a hybrid Tauc–Lorentz and Drude–Lorentz optical model. The retrieved dielectric tensor sheds light on another hyperbolic spectral region emerging below 341 nm, originating from anisotropic interband transitions. While this UV hyperbolic region is accompanied by strong optical losses, the Vis–NIR one observed above 509 nm exhibits comparatively low imaginary permittivity components and is driven by Drude-type free-carrier response along the metallic axis. Furthermore, leveraging the colossal in-plane anisotropy of vdW $MoOCl_2$, we show an emergence of twist-induced strong Vis-NIR chiral optical response in sub-wavelength vdW $MoOCl_2$ bilayers. Our proof-of-concept studies with 62° twisted bilayers of 48 nm and 58 nm show good agreement with the calculated 48 % circular dichroism in transmitted signal spectra, confirming the feasibility of vdW $MoOCl_2$-based ultrathin chiral photonic devices [4].

References

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Figures

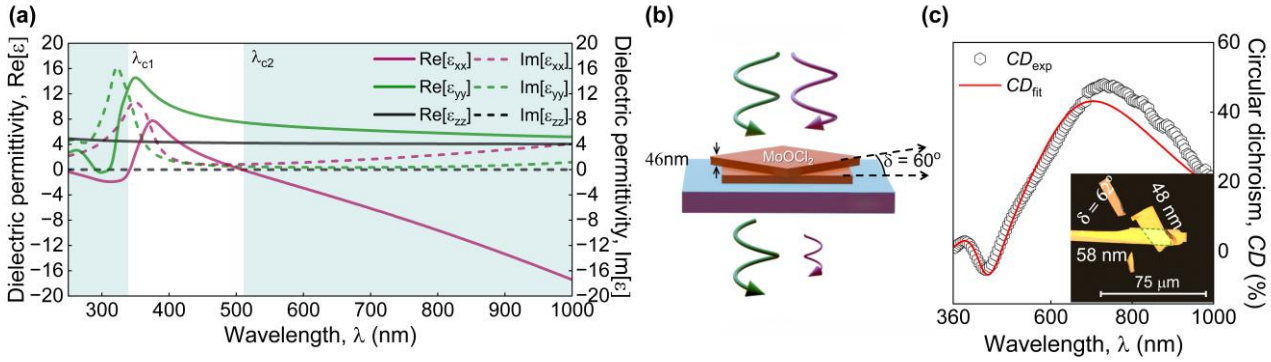

Fig. 1| The two UV and Vis-NIR hyperbolic regions of vdW $MoOCl_2$ and the twist-induced strong optical chirality in its sub-wavelength thick bilayers.