## Observation of the effect of surface chirality induced by chiral molecular overlayer on achiral substrates

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Symmetry is among the most important factors of materials, which governs their physical properties. For this reason, lowering of materials' symmetry by external manipulations has been actively utilized in exploring novel properties of matters, for example seen in twisted stacking or strain induction in layered materials, such as transition metal dichalcogenides, to name a few<sup>1</sup>.

In this regard, it is also well known that the materials' symmetry near the surface can readily be reduced by two-dimensional crystallization of *chiral* molecules on an achiral substrate<sup>2</sup> (surface chirality), which has been utilized for an enantio-selective catalysis in chiral chemistry. Furthermore, such a chiral-modified surface is regarded as an ideal platform to study chirality-induced spin selectivity (CISS), which has attracted a great deal of attention in recent years<sup>3</sup>. Thus, it is not only important to expand our abilities to control the chiral molecular crystallization, but also much deeper understanding of how such chirality interacts with the properties of the substrates is required to uncover the true, yet unknown, roles of chirality in two-dimensional materials.

In this study, an enantiopure chiral molecule thiadiazole-[9]helicene (TD[9]H) monolayer has been fabricated on Au(111), and probed using high-sensitivity low-energy electron diffraction (LEED) and scanning tunneling microscopy (STM) to study the potential landscape at their interface. Our analysis clearly revealed that TD[9]H molecules on Au(111) form an overlayer *without* commensurability, while still exhibiting substrate-defined surface chirality. Since the latter has only been reported in previous studies for the limited cases of commensurate overlayers, our findings serve to extend the possibility of fabricating well-defined surface chirality into a much broader spectrum of molecule-substrate combinations<sup>4</sup>.

Surface chirality was then formed on monolayser (ML)-WS<sub>2</sub>/Au(111) and ML- $Mo_{0.18}W_{0.82}S_2/Au(111)$ , hosts of highly spin-polarized electrons. We have successfully induced the surface chirality on the substrate, and the influence of the surface chirality was explored by measuring the band structure before and after the formation of surface chirality, using ARPES. Our results directly elucidate the changes in the band structure of the substrate, and indicate the chiral molecular adsorption likely induces an enhancement of the spin-polarized electron lifetime.

**Keywords:** Surface chirality, Low-energy electron diffraction, Angle-resolved photoelectron spectroscopy, Scanning tunneling microscopy

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