

Transition metal dichalcogenide surfaces as scattering targets in spin-polarization detectors: A case study of MoS₂

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The suitability of transition metal dichalcogenides (TMDCs) for the use as scattering targets in electron spin-polarization detectors is put to a paradigmatic test: a case study of MoS₂ is performed. Therefore, the electron reflectivity, the Sherman function, and the figure of merit are measured for the following samples: a single layer of MoS₂ on Au(111) and cleaved MoS₂ single-crystal surfaces. MoS₂ offers promising features for the use in spin-polarization detection: working points with a reversed sign of the Sherman function and a long-term target stability. To overcome the low figure of merit of MoS₂, the closely-related 2D material WS₂ with higher spin-orbit interaction is proposed, which may resemble the promising features with higher Sherman function. This study demonstrates the potential suitability of 2D materials such as TMDCs as scattering targets in spin-polarization detectors.

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I. INTRODUCTION

The spin polarization of electrons plays a crucial role in, e.g., photoemission experiments [1–3]. The spin texture of the electronic states under investigation is revealed by the use of spin-polarization detectors [4]. This leads to a deeper understanding of the investigated material and opens the way to possible applications in, for instance, the field of spintronics. Examples of spin-dependent electronic states are electronic states of ferromagnetic materials [1,5], surface states at high-*Z* materials [6], valence band maxima of 2D semiconductors [7,8], electronic states of complex, and layered systems [9].

The detection of electron spin polarization is based on spin-dependent electron scattering from targets influenced by exchange and/or spin-orbit interaction. One important characteristic of a spin-polarization detector is the efficiency of the spin separation, i.e., the figure of merit

$$\text{FOM} = \frac{I}{I_0} S^2. \quad (1)$$

The figure of merit depends on the ratio of the intensities of the scattered (*I*) and the incoming (*I*₀) electrons and predominantly on the square of the Sherman function *S* of the scattering target [4]. The Sherman function *S* describes the

spin-resolving power of the spin-polarization detector, which can be understood as the intensity asymmetry $(I_+ - I_-)/(I_+ + I_-)$ of a scattered electron beam with an initial spin polarization of *P* = 100%. It is described as polarization asymmetry factor as well [10].

Spin-polarization detectors of Mott [11] and SPLEED (spin-polarized low-energy electron diffraction) [12] type are based on spin-dependent electron scattering from targets influenced by spin-orbit interaction, e.g., a gold foil [13] and a W(001) surface [14], respectively. While Mott detectors are operated at higher scattering energies (≈10–100 keV), SPLEED detectors exploit the spin-dependent electron diffraction at more accessible, lower scattering energies (≈100 eV). Inherent to these types of detectors are the spin-dependent left-right and up-down scattering asymmetries, which enable to detect two spin-polarization components of the incoming electrons in one measurement. For instance, one component of the unknown spin polarization *P* of the incoming electrons is related to the Sherman function *S* of the detector by

$$P = \frac{I_{\text{left}} - I_{\text{right}}}{I_{\text{left}} + I_{\text{right}}} \frac{1}{S}. \quad (2)$$

Here, the relating factor is the intensity asymmetry of the two measured intensities *I*_{left} and *I*_{right}. Typical values of *S* and FOM range from 20% to 40% and 10^{−4} to 10^{−3}, respectively [13–16]. By using two orthogonally mounted detectors in one experimental setup, a three-dimensional spin-polarization analysis becomes feasible in two successive measurements [17].

A more recent type of spin-polarization detector is based on the very-low-energy electron diffraction (VLEED) process.

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These detectors rely on the spin-dependent reflection [(0,0) diffraction spot] of very-low-energy electrons (<30 eV). Two successive reflection measurements by using two working points of the target are necessary to detect one component of the spin polarization of the incoming electrons. Preferably, the two working points have a reversed sign of the Sherman function S and a similar efficiency FOM. Since the reflected intensity I , the Sherman function S , and the efficiency FOM vary with scattering energy and angle, a working point is defined by the scattering energy and angle. In the case of a target solely influenced by spin-orbit interaction, the sign reversal of the Sherman function S is usually achieved by a small change of the scattering energy. In the case of a ferromagnetic target, two opposite magnetization states can be used to obtain suitable working points.

It is noted that the terms VLEED and SPLEED are not uniquely used in the literature. The term VLEED is often used to describe the reflection of low-energy electrons from exchange-influenced surfaces (e.g., [18,19]) and the term SPLEED for reflection from spin-orbit-influenced surfaces (e.g., [20–22]). However, the electron reflection from exchange-influenced surfaces is termed SPLEED as well (e.g., [22]). Also, the (spin-integrated) electron reflection from high- Z surfaces is termed VLEED (e.g., [23,24]). In the present work, the term VLEED is used for very-low-energy electron reflection from both exchange- and spin-orbit-influenced surfaces. This definition avoids confusion with the SPLEED detector described above, which is based on left-right and up-down intensity asymmetries of higher-order diffraction and not on electron reflection.

A widely used VLEED spin-polarization detector [19,25–27] is based on spin-dependent electron reflection at a ferromagnetic surface [18,28–30]. Electrons with an unknown spin polarization P are reflected at the ferromagnetic surface, which is magnetized in two opposite directions ($\uparrow$ and $\downarrow$) for two successive measurements. The sign of the Sherman function S is reversed upon magnetization reversal. The unknown spin polarization P of the incoming electrons is related to the Sherman function S of the detector by

$$P = \frac{I_{\uparrow} - I_{\downarrow}}{I_{\uparrow} + I_{\downarrow}} \frac{1}{S}. \quad (3)$$

In analogy to before, the relating factor is the intensity asymmetry of the two measured intensities of the reflected electrons ($I_{\uparrow}$ and $I_{\downarrow}$) at the two opposite magnetization states of the ferromagnetic target. In the case of a passivated iron surface, scattering energies of about 6 and 13.5 eV are used. The reported values for S and FOM range from 20% to 40% and 2×10^{-3} to 1.9×10^{-2} , depending on the scattering energy [19,25–27]. By using two orthogonally mounted exchange-based VLEED detectors, a three-dimensional spin-polarization analysis becomes feasible [31] as in the case of Mott/SPLEED detectors described above.

Spin-polarization detection in modern photoemission experiments [32,33] becomes challenging, when several photoelectron energies and/or angles are detected simultaneously with, for example, a display-type analyzer. The resulting enhancement in efficiency is especially advantageous for surfaces, which degrade quickly over time. For these analyzers, a multichannel spin-polarization detector is needed, e.g.,

relying on the VLEED process [34–36], where the photoelectron distribution is scattered spin dependently at a target prior to detection. Thus, a spin-filtered image is measured. Although two successive measurements are needed to detect one component of the spin polarization, VLEED-based detectors are promising candidates for parallel detection. Alternative concepts, for example based on Mott scattering [37] or spin-dependent electron transmission through a ferromagnetic layer with subsequent electron-photon conversion in a semiconductor heterostructure [38], are proposed as well.

Regarding VLEED-based detectors in multichannel experiments, an oxygen-passivated iron surface [36,39] (predominantly influenced by exchange interaction) or W(001) [34,35,40], Ir(001) [41], and Au/Ir(001) [36,42,43] surfaces (influenced by spin-orbit interaction) were employed. The Au/Ir(001) target offers two suitable working points with Sherman functions S of -65% and 57% at scattering energies of 10.25 and 11.50 eV, respectively [42]. The two working points are characterized by a two-dimensional efficiency $\text{FOM}_{2D} = N \times \text{FOM}$ of 28 and 37, respectively. For $N = 5000$ data points [42], FOM values of 5.6×10^{-3} and 7.4×10^{-3} can be derived for the working points, respectively.

The working points of the Au/Ir(001) target described above are related to multiple electron scattering between the surface barrier and the crystal surface [43]. This especially occurs for electrons with energies few eV below the threshold energy of LEED beams and has the following reason. During the scattering process, the electrons can be diffracted. Since their energy is below the respective threshold energy, the electrons are trapped between the surface barrier and the crystal surface and are scattered back and forth multiple times [44]. Eventually, the electrons are backdiffracted to the specular direction. This mechanism can induce comparatively high absolute values of the Sherman function S , as in the case of the Au/Ir(001) target [43]. In addition, at energies below threshold, the electron reflectivity may have higher values as well, since no intensity is lost to the respective diffraction channel. This makes features in the electron reflectivity and Sherman function just below threshold energies of diffracted beams interesting in view of possible working points. Also above threshold, high values of the Sherman function S were observed for Au(111)/W(110) [45]. It is noted that the mechanism described above is responsible for VLEED fine structures, which are Rydberg-like series in the intensity and converge towards threshold energies of LEED beams [44,46–48].

To identify suitable working points, the spin-dependent electron reflectivity of a target must be known [43,49–52]. In a spin-polarized VLEED experiment, as performed in this work, the three important quantities electron reflectivity $R = I/I_0$, Sherman function S , and FOM are measured. Therefore, it is optimally suited for the search of new and efficient scattering targets for the use in spin-polarization detectors.

A promising target for the use in a spin-polarization detector has to fulfill criteria regarding (a) the working points, (b) the target stability and preparation procedure, and (c) the structural arrangement of the surface.

(a) The target has to offer at least two working points preferably with similar electron reflectivities R , similar absolute values of the Sherman function S , but with a reversed sign, and a high and similar efficiency of the spin separation

FOM. If these criteria are not exactly met, spin-polarization analysis is still feasible, but becomes more elaborate [40].

(b) The spin-dependent scattering characteristics need to be stable in UHV at least over the course of a measurement. In an optimal scenario, a target stability of several hours to days and longer is available. If the absolute value of the Sherman function S decreases over time, the determined spin polarizations become more and more unreliable. Furthermore, an easy and reproducible way of preparing the target is desirable.

(c) Regarding its use as a multichannel target, additional demands to the structural arrangement of the surface exist. A large-scale homogeneity of the surface on the mm-scale is required, whose size depends on the operational mode [41]. All electrons need to have the same scattering conditions. In that way, the beam characteristics do not change after the scattering process due to a rough or tessellated surface. A substantial widening of the electron beam after the scattering process would reduce the number of parallel resolvable data points. Furthermore, no superstructure or reconstruction at the surface is allowed. This leads in general to additional diffraction spots around a main diffraction spot. This circumstance makes it hard (or even impossible) to match the incoming electrons with the outgoing, spin-filtered electrons in the multichannel approach.

In this work, a spin-orbit-influenced transition metal dichalcogenide (TMDC) surface is put to a test regarding its applicability in a spin-polarization detector. Out of the many TMDCs, the prominent and widely-studied 2D material MoS₂ [53–55] is chosen for this paradigmatic test with spin-polarized VLEED. MoS₂ (like several other TMDCs) shows a high chemical inertness under UHV conditions. This is a striking advantage beforehand regarding the target stability. First, a single-layer (SL) MoS₂/Au(111) sample is investigated. SL MoS₂ samples on Au(111) are known to give valuable information about the material [56,57]. Second, cleaved surfaces of MoS₂ single crystals are investigated for reasons of comparison. Potential working points of MoS₂ will be discussed.

II. EXPERIMENTAL SETUP AND SAMPLE DETAILS

A. Spin-polarized VLEED setup

The experimental setup for spin-polarized VLEED is displayed in Fig. 1 [50,52,58]. The setup is located in an UHV apparatus with a base pressure of $p \approx 2 \times 10^{-10}$ mbar. Spin-polarized electrons from a GaAs photocathode [59–61] are guided onto the sample with varying electron energies E up to 30 eV with respect to the vacuum level E_V . The spin polarization of the electron source ($P = 33\%$) is reversed by changing the helicity of the laser light used for photoexcitation (σ^+ , σ^-). The spin-dependent intensities of the reflected electrons $I_\uparrow$ (for σ^+) and $I_\downarrow$ (for σ^-) are detected with a Faraday cup (detector). The polar and azimuthal electron angle of incidence Θ and φ , respectively, can be varied over a wide angular range. The position of the detector can be adjusted accordingly to maintain reflection geometry. The spot size of the electron beam on the sample is in the order of one to three millimeters.

After electrostatic deflection of the emitted electrons from the GaAs source in a 90° deflector, the spin polarization is

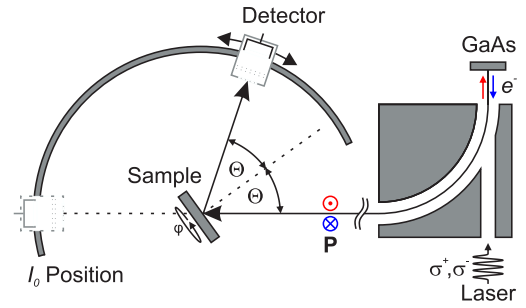


FIG. 1. Experimental setup for spin-polarized VLEED measurements. Spin-polarized electrons from the GaAs source are guided onto the sample with variable energy. The reflected electron intensities are measured with the detector. The electron reflectivity R , the Sherman function S , and the FOM of a sample can be measured over a wide range of incident electron energies and angles (adapted from Ref [52]).

transversal. The wavy lines in the sketch of the experimental setup indicate that the GaAs source is rotated by 90° with respect to the measuring plane. In that way, the spin polarization is perpendicular to the scattering plane and, thus, the electron orbital and spin moments are collinear for maximum sensitivity to spin-orbit-induced effects [4].

With this setup, the electron reflectivity $R = I/I_0$ (averaged over both spin channels), the spin-orbit-induced Sherman function $S = (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow) \times 1/P$ and the resulting figure of merit $FOM = R \times S^2$ are measured over a wide range of incident electron energies and angles for the SL MoS₂/Au(111) sample and cleaved MoS₂ single-crystal surfaces. The intensity I_0 is measured by moving the detector to the I_0 position with the sample retracted. Note that the electron reflectivities are displayed on a logarithmic scale in the figures, because they vary over orders of magnitude.

B. Preparation and characterization of the samples

SL MoS₂ was epitaxially grown on a clean and well-ordered Au(111) substrate. The growth of MoS₂ islands took place at 1×10^{-6} mbar chamber background pressure of dimethyldisulfide (DMDS, purity $\geq 99.0\%$) and a sample temperature of 750 °C. After 10 minutes of exposure to DMDS, to ensure full sulfidization, Mo was evaporated from a Mo metal rod (purity $\geq 99.95\%$). A total growth rate of 0.1 SL/h was calculated by the final coverage of 0.3 SL, which itself was determined by large-scale low-energy electron microscopy (LEEM) observations in relation to the evaporation time. A detailed description of the growth mechanism is given by Ewert *et al.* [62]. An incomplete adlayer coverage was chosen to avoid multilayer growth because, with these growth conditions, bilayer growth at the boundaries of the MoS₂ islands is about to start [63].

After transfer into the UHV apparatus in Münster, the SL MoS₂/Au(111) sample was annealed at temperatures of up to $T = 600$ °C to remove contaminants. After initial, soft annealing at $T = 180$ °C, the temperature was increased progressively up to $T = 600$ °C. Higher annealing temperatures than $T = 600$ °C were excluded to prevent possible MoS₂ decomposition. A LEED pattern of the annealed sample is

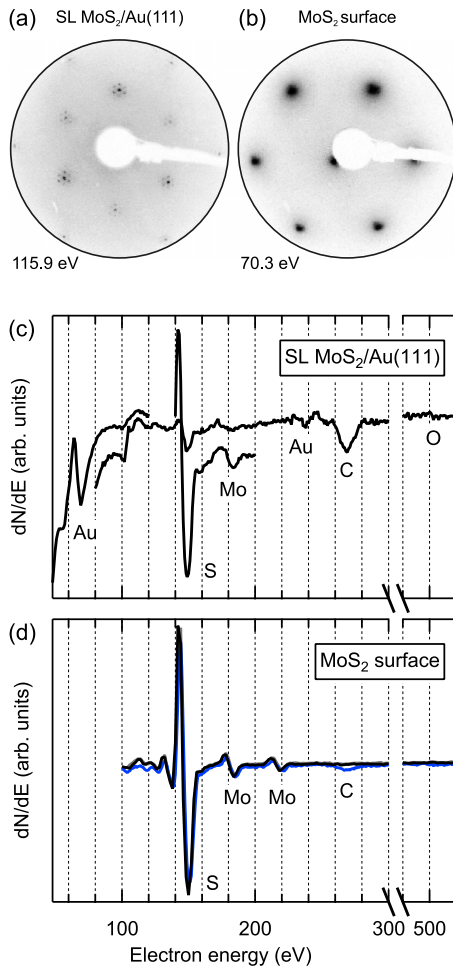


FIG. 2. LEED images of (a) the SL MoS₂/Au(111) sample (electron energy $E = 115.9$ eV, annealed at $T = 270^\circ\text{C}$) and (b) the surface of a MoS₂ crystal (electron energy $E = 70.3$ eV, cleaved in UHV). AES results (obtained with variable sensitivity of the detection system) of (c) the SL MoS₂/Au(111) sample (annealed at $T = 270^\circ\text{C}$) and (d) the surface of a MoS₂ single crystal (blue lines: cleaved in atmosphere, black lines: cleaved in UHV, gray lines (almost covered by the black lines): cleaved in UHV and stored for 8 weeks in UHV).

shown in Fig. 2(a). It shows diffraction spots typical for a SL MoS₂/Au(111) sample [7,64] including the moiré structure, which is induced by the lattice mismatch of SL MoS₂ and Au(111). Results of Auger electron spectroscopy (AES) in Fig. 2(c) show characteristic features of sulfur, molybdenum, and gold. In addition, a sizable carbon signal and a slight oxygen signal are observed, presumably due to contamination during sample transfer in atmosphere. We want to emphasize that annealing temperatures between $T = 180^\circ\text{C}$ and $T = 600^\circ\text{C}$ did not cause any significant changes in the VLEED signal. A comparison with VLEED measurements on a pristine and a contaminated Au(111) surface indicates that the carbon contamination of the single-layer sample is located on the uncovered area and not on the MoS₂ islands (see Supplemental Material [65]).

Surfaces of MoS₂ single crystals (2H polytype structure, p-doped) were cleaved in atmosphere and in UHV. In Fig. 2(d),

corresponding AES measurements are shown. The AES results of the MoS₂ surface cleaved in UHV show exclusively molybdenum and sulfur signals. In contrast, the AES results of the MoS₂ surface cleaved in atmosphere include an additional peak at ≈ 270 eV, which indicates carbon contamination. A LEED pattern of the MoS₂ surface cleaved in UHV is shown in Fig. 2(b). A similar LEED pattern was observed for the surface cleaved in atmosphere. In both cases, no indications of the expected three-fold symmetry of the 2H polytype structure were observed in the diffraction pattern. This is attributed to the electron beam used for the LEED experiment that averages over several terraces of the MoS₂ surface. The diffraction spots are rather broad. This is essentially due to the fact that the MoS₂ surface consists of stitched domains/facets, which enclose a small angle with each other. The broad shape of the diffraction spots may also be caused by islands on the nanoscale.

III. VLEED MAPS OF MoS₂

To identify suitable working points of MoS₂, the electron reflectivity R , the Sherman function S , and the resulting figure of merit FOM are investigated for the SL MoS₂/Au(111) sample and cleaved MoS₂ single-crystal surfaces over a wide range of incident electron energies and angles. A detailed comparison will show that each target has its own pros and cons regarding the preparation and the structural arrangement of the surface.

A. SL MoS₂/Au(111)

The SL MoS₂/Au(111) sample (30% adlayer coverage) offers atomically flat islands of single-layer MoS₂ and a refreshment of the surface only demands degassing at soft annealing temperatures [64]. The stability of the spin-dependent scattering characteristics of the SL MoS₂/Au(111) sample in this experiment has proven to be at least 2 weeks under UHV conditions ($p \approx 2 \times 10^{-10}$ mbar). The VLEED signals of the SL MoS₂/Au(111) sample, right after preparation (soft annealing) and after 2 weeks stored in UHV, were equivalent within the uncertainty of the measurement (data not shown here).

In Fig. 3, the experimental results of the SL MoS₂/Au(111) sample are displayed as polar maps (polar angle of incidence vs kinetic energy). The electron reflectivity R (top) has distinct features at very low electron energies with a maximum reflectivity of $\approx 5\%$. The features below 10 eV are pinned to the threshold energy of the first LEED beam (black, dashed line in the map) and, thus, are related to a VLEED fine structure [48]. The VLEED spectra show significant features below 20 eV in accordance with normal-incidence, spin-integrated electron reflectivity spectra recorded in LEEM on selected single-layer MoS₂ islands [62,63,66]. A direct comparison is not feasible because of the different electron angles of incidence in the two experiments.

The Sherman function S (mid) shows several sign reversals at very low electron energies. At angles of incidence $\Theta > 40^\circ$ and electron energies E below 5 eV one finds a sign reversal, which is quite insensitive to small changes of the angle of incidence. This feature offers maximum absolute S values of $\approx 5\%$. Additionally, it is located in the region of the

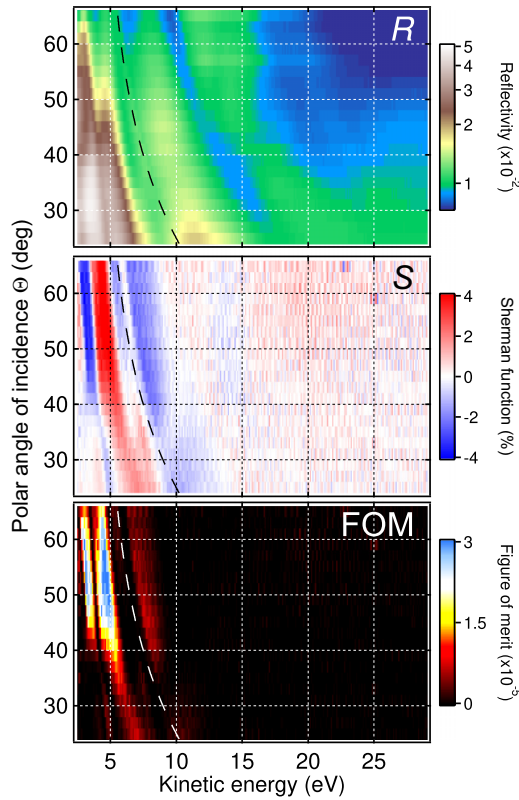


FIG. 3. Spin-polarized VLEED signal of the single-layer $\text{MoS}_2/\text{Au}(111)$ sample. For this measurement the single-layer sample was annealed at $T = 300^\circ\text{C}$. Electron reflectivity R (top), Sherman function S (mid), and figure of merit FOM (bottom) are displayed as polar maps (R on a logarithmic scale). The high symmetry direction $\overline{\Gamma}\text{M}$ lies in the scattering plane. The dashed lines in the polar maps display the threshold energy of the first LEED beam.

map with higher electron reflectivity R . Thus, the figure of merit FOM (bottom) at these angles and energies, or possibly working points, amounts to $\text{FOM} \approx 3 \times 10^{-5}$. Interestingly, the working points are located just below the threshold energy of a LEED beam, where relatively high absolute values of the Sherman function S in combination with higher electron reflectivity R are expected, as described above.

To survey the azimuthal dependence of the working points described above, the polar angle of incidence was set to $\Theta = 45^\circ$ and the sample was rotated around its azimuth. In Fig. 4, the result is displayed as an azimuth map (azimuth angle vs kinetic energy). By changing the azimuth from the $\overline{\Gamma}\text{M}$ direction to the $\overline{\Gamma}\text{K}$ direction, the aforementioned feature of the Sherman function S shifts to slightly higher electron energies. In the vicinity of the $\overline{\Gamma}\text{K}$ direction, the negative peak (blue) is largely suppressed, while the value of the positive peak (red) is only slightly enhanced. The suppression of the negative peak along $\overline{\Gamma}\text{K}$ promotes the use of the features of the Sherman function S along $\overline{\Gamma}\text{M}$ as possible working points in a spin-polarization detector. The features in the azimuth map around 8 to 10 eV offer relatively large negative values of the Sherman function S . However, these features do not offer the possibility to reverse the sign of the Sherman function S by a small change of the scattering energy and are, thus, not favorable for the use as working points.

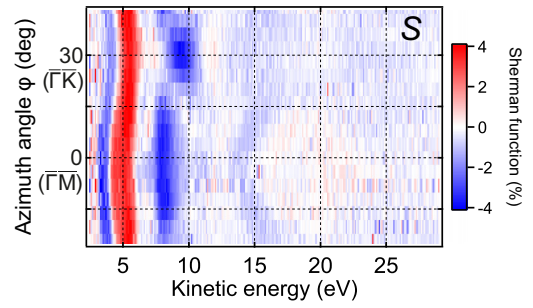


FIG. 4. Azimuthal dependence of the features in the Sherman function S of the single-layer $\text{MoS}_2/\text{Au}(111)$ sample. For this measurement the single-layer sample was annealed at $T = 180^\circ\text{C}$. The Sherman function S is displayed as an azimuth map. The polar angle of incidence was set to $\Theta = 45^\circ$.

Two major drawbacks of the SL $\text{MoS}_2/\text{Au}(111)$ sample regarding the use as a multichannel target are the incomplete adlayer coverage and the structural interaction of SL MoS_2 with the $\text{Au}(111)$ substrate (moiré structure) as discussed as criterion (c) in the introduction. The moiré structure leads to satellite spots around each diffraction spot [see Fig. 2(a)]. This includes the (0,0) spot, i.e., the reflected electron beam, which is relevant for VLEED.

B. MoS_2 surface

The drawbacks of the SL $\text{MoS}_2/\text{Au}(111)$ sample are overcome by the MoS_2 single-crystal surface, which provides a full surface coverage of MoS_2 and shows no moiré pattern. For preparation, this surface needs only to be cleaved.

In Fig. 5, the electron reflectivity R (top), the Sherman function S (mid), and the figure of merit FOM (bottom) of the MoS_2 single-crystal surface cleaved in atmosphere (a) and in UHV (b) are displayed as polar maps. In both cases, R and S have qualitatively the same structures. Quantitatively, the values of R and the absolute values of S are reduced in the case of the MoS_2 surface cleaved in atmosphere. This behavior is reflected in the FOM (bottom): the FOM of the MoS_2 surface cleaved in UHV ($\approx 10^{-4}$) is one order of magnitude larger than the FOM of the MoS_2 surface cleaved in atmosphere ($\approx 10^{-5}$). This reduction is attributed to the carbon contamination of the MoS_2 surface cleaved in atmosphere. A reduction of the reflected electron intensity in the case of adsorbates on top of bilayer MoS_2 was also observed in spin-integrated LEEM experiments [67]. The qualitative comparability of the results in Figs. 5(a) and 5(b), however, confirms the well-known robustness of MoS_2 (e.g., [68,69]). In addition, it shows the reproducibility of the results for different cleaves, since the qualitative VLEED signatures of MoS_2 are preserved. It is noted that the VLEED reflectivity of the MoS_2 surface cleaved in UHV shows similar structures as recently published ARRES (angle-resolved reflected electron spectroscopy) results of a MoS_2 surface [70].

By comparing the results of the cleaved MoS_2 single-crystal surfaces (see Fig. 5) and the SL $\text{MoS}_2/\text{Au}(111)$ sample (see Fig. 3), a high similarity is apparent. This shows that the uncovered area qualitatively has a rather low impact on the signal of the SL $\text{MoS}_2/\text{Au}(111)$ sample. The negligible

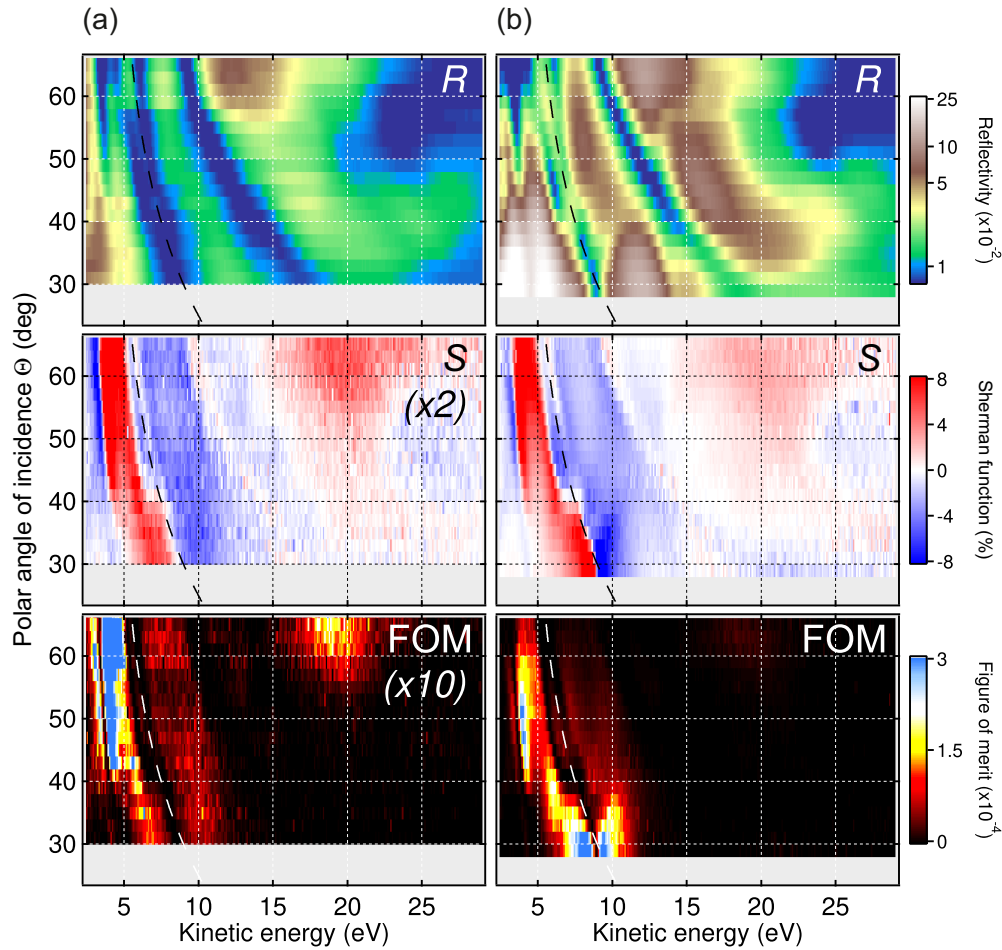


FIG. 5. Spin-polarized VLEED signal of the MoS₂ surface for two different surface conditions (a) and (b). For (a), the MoS₂ surface was cleaved in atmosphere. A carbon signature was observed in corresponding AES measurements. For (b), the MoS₂ surface was cleaved in UHV. No signatures of any adsorbates were observed in corresponding AES measurements. Electron reflectivity R (top), Sherman function S (mid), and figure of merit FOM (bottom) are displayed as polar maps (R on a logarithmic scale). The high symmetry direction $\bar{\Gamma}-\bar{M}$ lies in the scattering plane. The dashed lines in the polar maps display the threshold energy of the first LEED beam.

influence of the uncovered area of the single-layer sample is demonstrated by a comparison with VLEED signals of a contaminated and a pristine Au(111) surface (see Supplemental Material [65]).

Quantitatively, the values of R and the absolute values of S of the SL MoS₂/Au(111) sample are in general lower in comparison with the MoS₂ surface (valid for both surface conditions). This observation is in line with the fact that the SL MoS₂/Au(111) sample offers less MoS₂ since the adlayer coverage only amounts to $\approx 30\%$. Moreover, it can be attributed to the diffuse electron scattering of the uncovered area.

The high similarity of R and S of the cleaved MoS₂ surfaces and the SL MoS₂/Au(111) sample leads consequently to similar structures of the FOM (bottom row in Figs. 3 and 5). The identified working points of the SL MoS₂/Au(111) sample are also observed in the case of the MoS₂ surface. These consist of the sign reversal of S in regions of higher R values for polar angles of incidence $\Theta > 40^\circ$ and energies few eV below the threshold energy of the first LEED beam.

An additional set of promising working points with $\text{FOM} \approx 3 \times 10^{-4}$ arises for the MoS₂ surface cleaved in UHV for polar angles of incidence $\Theta \leq 31^\circ$ and energies between 6 and 11 eV. These working points are only weakly pronounced in the case of the MoS₂ surface cleaved in atmosphere and the SL MoS₂/Au(111) sample. The latter features are located in the vicinity of the threshold energy of the first LEED beam as well.

The FOM of the MoS₂ surface cleaved in UHV ($\text{FOM} \approx 10^{-4}$) is in general one order of magnitude larger than the FOM of the SL MoS₂/Au(111) sample ($\text{FOM} \approx 10^{-5}$) grown in a separate UHV chamber and transported in atmosphere.

The major drawback of the cleaved MoS₂ single-crystal surface regarding its use as a multichannel target in a spin-polarization detector is the poor structural/topographic quality of its surface. This is reflected in the LEED pattern of the MoS₂ surfaces [see Fig. 2(b)] and was already discussed. Additionally, the topography of the surface is in general different after each cleave.

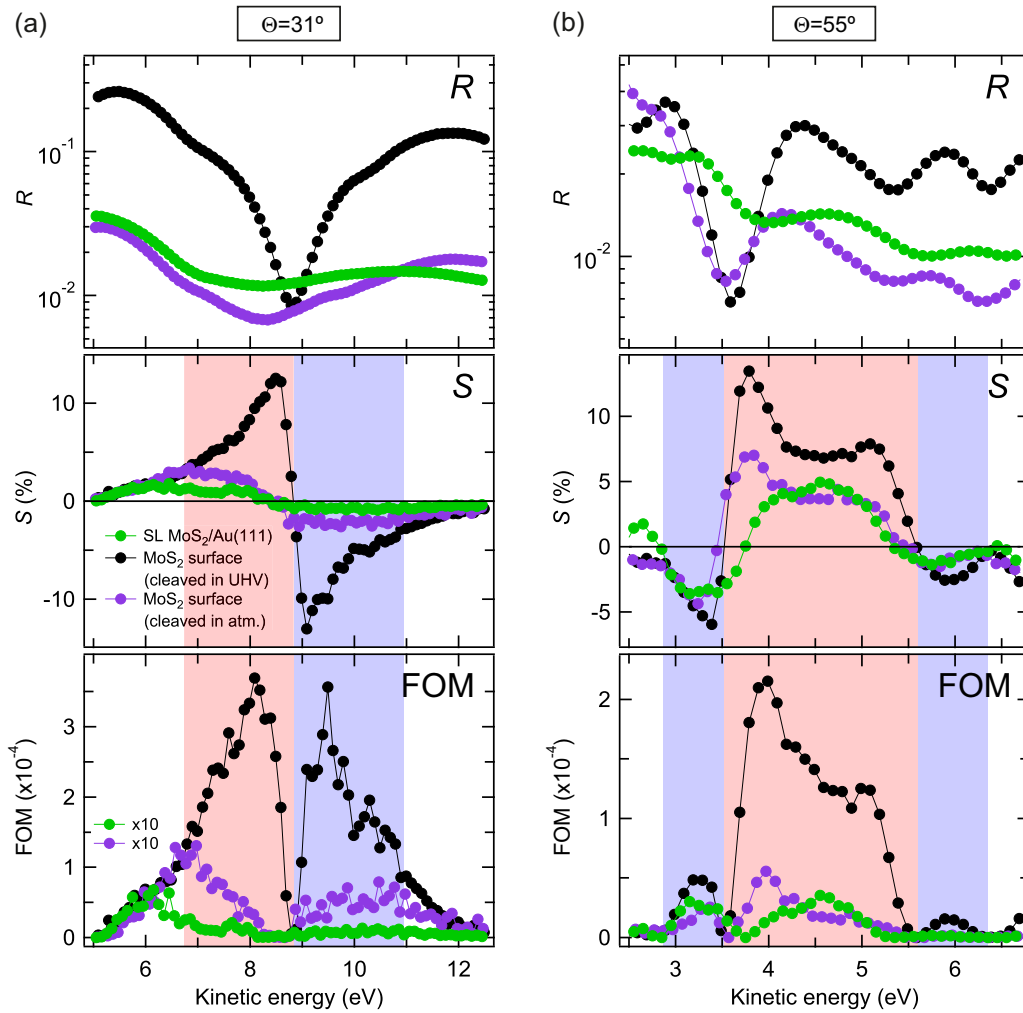


FIG. 6. Identified working points of MoS₂ exemplarily displayed for polar angles of incidence of $\Theta = 31^\circ$ [in (a)] and $\Theta = 55^\circ$ [in (b)], respectively. Electron reflectivity R (top row, on a logarithmic scale), Sherman function S (middle row), and the figure of merit FOM (bottom row) are displayed in the corresponding energy range for the SL MoS₂/Au(111) sample (green dots) and the MoS₂ surface (purple dots: cleaved in atmosphere, black dots: cleaved in UHV). The areas marked in red (blue) highlight working points with a positive (negative) Sherman function S .

C. Working points of MoS₂

Considering the experimental results of both the SL MoS₂/Au(111) sample and the cleaved MoS₂ single-crystal surfaces, promising working points of MoS₂ were identified. These are located in Θ - E areas in the vicinity of the threshold energy of the first LEED beam. In Fig. 6, working points are exemplarily displayed for polar angles of incidence of $\Theta = 31^\circ$ and $\Theta = 55^\circ$. The electron reflectivity R (on a logarithmic scale), the Sherman function S , and the figure of merit FOM are displayed in the upper, middle, and bottom row, respectively.

Qualitatively, the working points of the three samples, SL MoS₂/Au(111) and the MoS₂ surfaces cleaved in UHV and in atmosphere, have similar features in R , S , and FOM. Quantitatively, the MoS₂ surface cleaved in UHV offers the highest (absolute) R , S , and FOM values. Therefore, the working points are discussed in view of the results of the latter.

For $\Theta = 31^\circ$, two working points with pronounced features in S and FOM are observed (marked by the red and

blue areas). At energies of, for instance, $E_1 = 8.2$ eV and $E_2 = 9.4$ eV, the two working points have R and S values of $R_1 = 3.4\%$ and $S_1 = +10.2\%$ as well as $R_2 = 3.0\%$ and $S_2 = -9.9\%$, respectively. This results in FOM values of $\text{FOM}_1 = 3.5 \times 10^{-4}$ and $\text{FOM}_2 = 2.9 \times 10^{-4}$. These are the most promising working points of the MoS₂ surface identified in this work.

For $\Theta = 55^\circ$, several working points with a reversed sign of S are observed (marked by the red and blue areas). However, they provide smaller (absolute) values of S and FOM than the working points for $\Theta = 31^\circ$. Also, the FOM values show rather large differences for different energies. For example, two working points with energies of $E_1 = 3.2$ eV and $E_2 = 4.0$ eV have R and S values of $R_1 = 2.4\%$ and $S_1 = -4.5\%$ as well as $R_2 = 1.9\%$ and $S_2 = +10.6\%$, respectively. This results in FOM values of $\text{FOM}_1 = 4.9 \times 10^{-5}$ and $\text{FOM}_2 = 2.1 \times 10^{-4}$. The FOM values differ by one order of magnitude. Similar FOM values for both spin channels are more suitable.

IV. EVALUATION OF TMDC SURFACES AS SCATTERING TARGETS

Comprehensive information about the spin-dependent scattering characteristics of MoS₂ was obtained by investigating a SL MoS₂/Au(111) sample and cleaved MoS₂ single-crystal surfaces. On this basis, the applicability of MoS₂ and further TMDCs for the use as a multichannel target in a spin-polarization detector is evaluated. The following discussion follows the criteria (a)–(c), which were formulated in the introduction.

(a) In Fig. 6, several working points of MoS₂ with a reversed sign of the Sherman function S are summarized. A clean MoS₂ surface offers a figure of merit of up to $\text{FOM} \approx 3 \times 10^{-4}$. This efficiency of the spin separation has the same order of magnitude as the efficiency of Mott detectors [11,13,16] and is one order of magnitude lower in comparison with state-of-the-art spin polarization detectors ($\text{FOM} \approx 10^{-3}$) [25,27,34–36,42], which are based on high- Z targets in most cases. By replacing, e.g., MoS₂ ($Z(\text{Mo}) = 42$) by WS₂ ($Z(\text{W}) = 74$) the absolute values of the Sherman function S and, thus, the figure of merit FOM are expected to rise. Additionally, WS₂ may keep the promising working points of MoS₂ due to the similar structure in comparison with MoS₂.

(b) The target stability of the SL MoS₂/Au(111) sample was tested in our apparatus and, as described above, amounts to at least 2 weeks under UHV conditions (data not shown here), which is comparable to the stability of the Au/Ir(001) scattering target [42]. A possible refreshment of the sample only demands soft annealing. Regarding the MoS₂ surface, the chemical composition was checked by an AES measurement several weeks after cleaving the MoS₂ single crystal in UHV. No AES signatures of carbon or any other adsorbates were observed [see Fig. 2(d)]. Based on these observations, a high target stability of MoS₂ in UHV can be assumed.

The process to cleave a TMDC bulk crystal in UHV is an easy exercise. The epitaxial growth of a TMDC on a given substrate is, however, a specialist's method. The epitaxially grown TMDC adlayer on a substrate can be transported under UHV conditions (for instance with a vacuum suitcase) to circumvent a possible carbon contamination after fabrication.

(c) Ultimately, a surface with a long-range homogeneity without any reconstruction or structural interaction with the substrate is needed regarding the use as a multichannel target. As described above, the SL MoS₂/Au(111) sample has cons regarding the incomplete adlayer coverage and the structural interaction of the adlayer with the substrate (moiré period-

icity). While the MoS₂ surface overcomes the drawbacks of the SL MoS₂/Au(111) sample, it consists of stitched domains/facets, which enclose a small angle with each other. A flat surface on the mm-scale is needed. SL WS₂/Au(111) seems suitable at first glance, but a moiré periodicity is present in this system, too [71,72]. To test epitaxially grown, large-area SL MoS₂ [73–75] and WS₂ [76] on sapphire seems promising. LEED images of these systems indicate that no moiré periodicity is present. Alternatively, epitaxially grown, large-area SL WS₂ on a gold foil [77] or mechanically exfoliated, large-area SL MoS₂ (and further TMDCs) put on polycrystalline gold [78] could be chosen. The growth of large-area bilayer MoS₂ (or possibly other TMDCs) on sapphire also seem promising [79]. The growth and/or production of these materials on the wafer scale is an on-going field of research. On the basis of the findings of this work, an investigation of the spin-dependent scattering characteristics of a full-coverage and atomically flat single- or bilayer TMDC with high- Z constituents (e.g., WS₂) seems promising.

V. SUMMARY

In this work, we performed a systematic spin-polarized VLEED study centered around the TMDC MoS₂. The electron reflectivity R , the Sherman function S , and the figure of merit FOM of the SL MoS₂/Au(111) sample and cleaved MoS₂ single-crystal surfaces were measured over a wide range of incident electron scattering energies and angles. This paradigmatic study of MoS₂ revealed the potential usefulness of TMDCs as scattering targets in spin-polarization detectors. Promising working points of MoS₂ were identified and a proposal for optimized TMDC-based scattering targets was given.

Concept of the study: C.A. and M.D. Measurements on the SL MoS₂/Au(111) sample: C.A. and N.E. Measurements on the cleaved MoS₂ single-crystal surfaces: C.A. and A.H. Measurements on the Au(111) surface (See Supplemental Material [65]): C.A. and A.R. Fabrication of the SL MoS₂/Au(111) sample: M.E., L.B., J.F., and J.I.F. Visualization: C.A. Writing - original draft: C.A. and M.D. Writing - review: C.A., A.H., N.E., A.R., M.E., L.B., J.F., J.I.F., and M.D. Supervision: M.D.

DATA AVAILABILITY

The data that support the findings of this article are available from the authors upon reasonable request.

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