

## Image-type very-low-energy-electron-diffraction spin polarimeter by 90-degree reflection

**Authors:** Zhang, Mingzhu, Wu, Muyang, Dr. Bo Zhao, Qiao, Dr. Shan, Dr. Bo Zhao

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### Abstract

The invention of multi-channel spin polarimeters has significantly promoted the spin- and angle-resolved photoelectron spectroscopy (SARPES) field. However, the current image-type multi-channel spin polarimeters suffer from their inability to measure all components of spin polarization. Here, we report a new image-type very-low-energy-electron-diffraction (VLEED) spin polarimeter with simpler structure, the potential to measure three-dimensional spin polarization and ease of adjustment, which utilizes the iron target as part of the electron optics to separate the paths of incident and reflected electrons by a 90-degree reflection. A SARPES station equipped with this polarimeter was developed at the 07U beamline of Shanghai Synchrotron Radiation Facility (SSRF). Its energy and angular resolutions achieve 5.3 meV and  $0.27^\circ$ , respectively. The single-channel efficiency of the spin polarimeter was verified through SARPES measurements for Bi (111) film, yielding a value of  $2.1 \times 10^{-2}$ .

### Full Text

### Preamble

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Ming-Zhu Zhang,<sup>1,2</sup> Mu-Yang Wu,<sup>1,2</sup> Bo Zhao,<sup>3,†</sup> and Shan Qiao<sup>1,2,4,‡</sup>

<sup>1</sup>National Key Laboratory of Materials for Integrated Circuits, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai 200050, China

<sup>2</sup>Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

<sup>3</sup>Shanghai Synchrotron Radiation Facility, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201204, China

<sup>4</sup>School of Physical Science and Technology, ShanghaiTech University, Shanghai 201210, China

The development of multi-channel spin polarimeters has significantly advanced the field of spin- and angle-resolved photoelectron spectroscopy (SARPES). However, existing image-type multi-channel spin polarimeters are limited by their inability to measure all components of spin polarization simultaneously. Here we report a new image-type very-low-energy-electron-diffraction (VLEED) spin polarimeter that addresses this limitation through a simpler structural design enabling three-dimensional spin polarization measurements with ease of adjustment. Our approach utilizes an iron target as an integral component of the electron optics to separate incident and reflected electron paths via 90-degree reflection. A SARPES station equipped with this polarimeter has been developed at the BL07U beamline of the Shanghai Synchrotron Radiation Facility (SSRF), achieving energy and angular resolutions of 5.3 meV and  $0.27^\circ$ , respectively. The single-channel efficiency of the spin polarimeter was verified through SARPES measurements on a Bi(111) film, yielding a value of  $2.1 \times 10^{-2}$ .

**Keywords:** spin- and angle-resolved photoelectron spectroscopy, spin polarimeter, band structure

## Introduction

Quantum materials represent a major research frontier in condensed matter physics due to their diverse and novel properties, including charge density waves [?], superconductivity [?, ?], topological surface states [?, ?, ?], spin density waves, and magnetism [?, ?, ?, ?]. These properties are intimately connected to their electronic structures, making spectroscopic measurements essential for understanding the underlying physical mechanisms, particularly the spin polarization arising from spin-orbit coupling or exchange interactions.

SARPES typically employs a hemispherical analyzer to measure the energy and momentum of photoelectrons, followed by a spin polarimeter to analyze their spin orientation. However, early Mott-type spin polarimeters based on spin-orbit interaction suffered from extremely low efficiency ( $\sim 10^{-4}$ ) [?, ?, ?, ?, ?], requiring  $10^4$  times longer acquisition times to achieve the same statistical accuracy as measurements without spin analysis. This limitation severely hindered SARPES applications. To overcome this challenge, very-low-energy-electron-diffraction (VLEED) spin polarimeters based on exchange interaction were developed [?, ?]. In these devices, low-kinetic-energy electrons reflect from a magnetic target, and spin polarization is determined from the asymmetry in reflectivities between opposite magnetizations of the iron target according to the relation  $P = A/S$ , where P, A, and S represent polarization, asymmetry, and Sherman function, respectively. The Sherman function characterizes the inherent asymmetry that would be observed for 100% polarized electrons.

VLEED spin polarimeters achieve efficiencies of approximately  $10^{-2}$ , representing a two-order-of-magnitude improvement over Mott-type devices [?, ?, ?, ?, ?].

To further enhance total efficiency through multi-channel detection compatible with modern image-type analyzers, several multi-channel spin polarimeters based on spin-orbit scattering from high-Z targets were developed, achieving single-channel efficiencies of  $1.7 \times 10^{-3}$  and  $6.4 \times 10^{-3}$ , respectively [?, ?]. Our group previously developed a multi-channel VLEED spin polarimeter based on exchange interaction with a higher single-channel efficiency of  $2.2 \times 10^{-2}$  [?, ?], which has proven particularly valuable for studying altermagnetic materials where simultaneous measurement of spin polarization symmetry across broad momentum regions is crucial [?, ?, ?].

For magnetic targets, a small Zeeman splitting exists between majority and minority spin bands, leading to significant momentum transfer during spin-flip scattering processes involving Stoner excitations [?]. This momentum transfer alters the exit angle relative to the incident angle, potentially disrupting electron optics functionality. Consequently, it was believed that the 90-degree reflection electron optics employed in spin-orbit-based multi-channel polarimeters would not function when the nonmagnetic target was replaced with a ferromagnetic material. The electron optics were thought to require normal incidence with coincidence between the electron intensity image plane and the target surface to maintain functionality [?]. To achieve normal incidence, our previous design incorporated a magnetic field, which complicated the electron optics and introduced significant aberrations due to edge effects. Although a second-generation design using permanent magnets with smaller gaps mitigated these issues and improved resolution [?], the fundamental problems of complex structure and large aberrations persisted. Moreover, the combination of electric and magnetic fields made optical design and alignment challenging.

A critical limitation of current multi-channel spin polarimeters is their inability to measure all three components of spin polarization. Existing polarimeters based on either spin-orbit or exchange interaction can measure only one or two spin components—either normal to the scattering plane or along the target surface. The VLEED spin polarimeter based on 90-degree reflection offers a potential solution to this limitation, prompting us to investigate whether such a design could operate without normal incidence. We discovered that Stoner excitations have negligible impact on VLEED polarimeters with 90-degree reflection. The working principle is illustrated in Fig. 1 Figure 1: see original paper, where the reflecting target becomes an integral part of the electron optics. The image at the hemispherical analyzer (HA) exit is transferred point-to-point by the first lens (Lens 1) to an image plane crossing the target center. This image plane is then transferred to the object plane of the second lens (Lens 2) via 90-degree reflection, which subsequently transfers it point-to-point to detector 2. Consequently, each point on detector 2 uniquely corresponds to a specific point on the HA exit plane. Since all object and image planes for both lenses are perpendicular to their optical axes, electron optical aberrations are minimized. The spin-integrated normal spectrum can be easily obtained by simply removing the target and recording electrons with detector 1 positioned behind it.

For a spin polarimeter using two lenses based on this principle (Fig. 1(a)), the spin polarization  $P_Y$  along the Y-direction can be obtained from the difference in electron intensities with magnetizations along positive and negative Y-directions. When the target is magnetized positively and negatively along the XZ-direction (perpendicular to both the Y-axis and the target normal), the spin polarization  $P_{\{XZ\}}$  along this direction can be measured. For a polarimeter with two perpendicular reflection arms (Fig. 1(b)), electrons can be directed to either detector 2 or detector 3 by rotating the target. After rotating the target to reflect electrons toward detector 3, the spin polarization along the X-direction can be obtained from intensity differences between positive and negative X-direction magnetizations. When the target is magnetized along the YZ-direction (perpendicular to both the X-axis and target normal), the spin polarization  $P_{\{YZ\}}$  along this direction can be measured. The spin polarization  $P_Z$  along the Z-direction can then be solved using the following equations:

$$\begin{aligned} P_{\{XZ\}} &= P_X \cos 45^\circ - P_Z \cos 45^\circ \\ P_{\{YZ\}} &= P_Y \cos 45^\circ - P_Z \cos 45^\circ \end{aligned}$$

which yields:

$$P_Z = (P_X + P_Y - (P_{\{XZ\}} + P_{\{YZ\}})) / 2$$

**Fig. 1.** (Color online) (a) Geometry of the electron optics for the multi-channel VLEED spin polarimeter by 90-degree reflection. (b) Schematic of the spin polarimeter capable of measuring three components of spin polarization.

## II. Description of BL07U SARPES Station

We now describe a new multi-channel VLEED spin polarimeter based on 90° reflection installed at the SARPES station of the BL07U beamline at SSRF. During construction of the SARPES station, the viability of the 90-degree reflection mode was uncertain, so a polarimeter with only one reflection arm was initially built for testing.

Figure 2: see original paper shows the overall layout of the SARPES station at the BL07U beamline of SSRF, comprising the analysis chamber, Scienta Omicron DA30L hemispherical analyzer, spin polarimeter, target preparation chamber, load-lock chamber, sample bank chamber, and sample preparation chamber. All chambers are interconnected under ultra-high vacuum conditions. The spectrometer is equipped with both an Xe discharge lamp providing 8.43 eV photons and synchrotron radiation spanning 50–2000 eV [?, ?]. The analysis chamber features a home-made six-degree-of-freedom manipulator with liquid helium cryostat. The spin polarimeter employs an Fe(001)-p(1×\$1) film terminated by oxygen (Fe(001)-O) as the reflection target, grown on an MgO(001) substrate in the target preparation chamber [?].

Figure 2(b) illustrates the spectrometer schematic. Photoelectrons emitted from the sample surface at different angles enter the HA at different slit positions.

The HA separates photoelectrons by kinetic energy into distinct positions, forming a two-dimensional image on its exit plane that serves as the source image for the spin polarimeter. To measure spin, a home-made image-type multi-channel VLEED spin polarimeter based on 90-degree reflection is installed. Electrons exiting the HA are focused onto the iron target by the first lens, undergo 90-degree reflection, and are refocused onto detector 2 by the second lens to record spin-resolved two-dimensional spectra. The spin-integrated angle-resolved photoelectron spectrum (ARPES) can be measured by detector 1 after target removal.

The detailed spin polarimeter structure is presented in Fig. 3 [Figure 3: see original paper]. Lens 1 and Lens 2 comprise multiple electrostatic cylindrical lenses and quadrupole lenses (formed by quartering cylindrical lenses), which transport electrons from the HA exit aperture to either detector in point-to-point focus mode. Cylindrical lenses control beam focusing and regulate image magnification at the detector plane, while quadrupole lenses deflect electrons to compensate for background magnetic field effects and lens alignment errors. All lens elements are electrostatically controlled through individual voltages supplied by dedicated power supplies. To facilitate rapid lens voltage adjustment, we developed control software enabling quick reading and storage of lens tables and fast switching between different lens tables via single-command operation, thereby enhancing experimental efficiency. The software also monitors voltage changes in real time and plots voltage variation curves to facilitate stability assessment.

To improve the signal-to-noise ratio of spin polarization measurements, multiple measurement loops are performed with the iron target magnetized in a positive-negative-negative-positive sequence. This magnetization pattern eliminates experimental errors arising from photoelectron intensity changes due to sample surface contamination over time. Automated target magnetization software was developed to streamline SARPES acquisition, enabling magnetization along the X-direction, Y-direction, or alternating between both to obtain spin polarization components independently or simultaneously. Communication between our home-made software and the SES software controlling the Scienta Omicron electron analyzer is essential. Our software monitors a designated data directory and, upon detecting newly generated spectral data, sends a command to generate a Ctrl-G event, prompting the operating system to communicate with SES and initiate a new spectral acquisition task. This ensures proper timing synchronization between magnetization and spectrum acquisition. When the preset number of measurement loops is completed, the software terminates acquisition and alerts the researcher, recording each magnetization direction in a timestamped log file.

**Fig. 3.** (Color online) Detailed structure of the spin polarimeter. The red line indicates the electron trajectory.

A single-crystal Bi(111) thin film grown on Si(111) serves as a standard sample for evaluating spin polarimeter performance [?]. High-quality Bi(111) growth

requires excellent vacuum in the growth chamber and a clean Si(111) substrate. Substrate cleaning is performed by flashing the silicon wafer to remove the surface oxide layer at a peak temperature of 1200°C while preventing wafer melting. Current magnitude, duration, and chamber vacuum level are critical for obtaining an atomically clean surface exhibiting a clear  $7\sqrt{3}\times 7\sqrt{3}$  low-energy electron diffraction (LEED) reconstruction pattern. We developed control software to regulate current parameters and monitor pressure changes during each flash cycle, ensuring the chamber reaches a preset pressure before automated flashing. For the SARPES station, vacuum level is critical for sample growth, processing, and measurement, making continuous vacuum monitoring essential for normal operations. We therefore developed real-time multi-chamber vacuum monitoring software that provides continuous vacuum level surveillance and alerts when pressures exceed preset thresholds.

### III. Results and Discussion

To evaluate spin polarimeter performance, we prepared a Bi(111) film by molecular beam epitaxy in the preparation chamber and confirmed its monocrystalline quality through LEED observation [?, ?]. The film was transferred in situ to the analysis chamber. Figure 4 Figure 4: see original paper shows the Fermi surface map of Bi(111) measured in ARPES mode using 8.43 eV photons from an Xe lamp. The central region reveals a hexagonal electron pocket accompanied by six hole pockets exhibiting threefold symmetry, consisting of three bright and three dark regions. This Fermi surface symmetry likely originates from the threefold symmetry of bulk Bi(111) crystals and reflects the Rashba splitting of the Bi(111) surface [?]. We also measured the electron band dispersion along the dashed line in Fig. 4(a), shown in Fig. 4(b). Rashba surface states near the Fermi level are clearly visible, consistent with the Fermi surface map. Two high-intensity bulk bands appear around 0.2 eV and 0.9 eV binding energy.

The scattering asymmetries of SARPES for surface states with binding energies from 0.6-0.8 eV at 3.5 eV scattering energy are shown in Fig. 4(c). The averaged momentum distribution curve (MDC) within the black-dashed square region of Fig. 4(c) is presented in Fig. 4(d). The averaged asymmetries are 0.39 and -0.24 near  $-0.04 \text{ \AA}^{-1}$  and  $0.04 \text{ \AA}^{-1}$ , respectively. In principle, the two split bands should exhibit 100% polarization with identical absolute asymmetries. The observed differences arise from transition matrix element asymmetries, allowing us to use 0.39 as a conservative estimate of the Sherman function, which is smaller than the true value since electron polarization cannot exceed 100%.

The Sherman function represents the asymmetry observed for 100% polarized electrons and depends only on the reflection target. However, when estimated from SARPES measurements of a standard sample, finite energy analyzer resolution mixes 100% polarized bands with opposite spin polarization, reducing the observed polarization below 100%. This leads to underestimation of the Sherman function, with experimentally estimated values increasing as angular and energy resolutions improve.

Measuring absolute reflectivity is challenging because electrons recorded by charge-coupled devices (CCDs) behind detectors 1 and 2 undergo two-stage multiplication through microchannel plates (MCPs) with unequal gains. To address this, we performed direct electron counting, where gain variations affect only pulse amplitude. Figure 5 Figure 5: see original paper schematically illustrates the experimental setup. Each electron entering the detector (comprising two MCPs coupled to a phosphor screen) generates a multiplied electron bunch striking the screen. An Ortec 142IH charge-sensitive preamplifier connected to the screen outputs a pulse with amplitude proportional to the total charge, which is recorded by an oscilloscope. Pulses are counted when their amplitudes exceed a threshold.

To determine the threshold for filtering electronic noise, we recorded signals with the analyzer slit closed, as shown in Fig. 5(b). High-amplitude pulses originate from cosmic rays penetrating the analyzer chamber, while low-amplitude pulses represent noise. Figure 5(c) shows average pulse counts over 100 seconds as a function of threshold amplitude. Noise counts decrease more rapidly with increasing threshold than signal counts. Linear fitting in logarithmic coordinates reveals an intersection at approximately 60 mV, establishing this as the optimal threshold for detector 1. This reliability is confirmed in Fig. 5(b). Signals recorded with an open analyzer slit are shown in Fig. 5(d). To estimate random error, we performed 100 one-second measurements, yielding an average total electron count  $N_{\{t1\}} = 724 \pm 27$  counts/s from detector 1 and average background cosmic ray count  $N_{\{c1\}} = 18 \pm 5$  counts/s. The photoelectron-induced count  $N_{-1} = N_{\{t1\}} - N_{\{c1\}}$  is therefore  $706 \pm 27$  counts/s.

The same method measured scattered photoelectron counts at detector 2, with an optimal threshold of 55 mV. At 5.0 eV scattering kinetic energy, total count  $N_{\{t2\}} = 214 \pm 18$  and background count  $N_{\{c2\}} = 18 \pm 5$  gave  $N_{-2} = 196 \pm 19$ , yielding a reflectivity  $R = 0.28 \pm 0.03$ . Measured reflectivities at various scattering kinetic energies are shown in Fig. 4(e).

Spin polarimeter efficiency serves as a crucial performance metric [?], with higher efficiency enabling faster acquisition of spin-resolved spectra. To optimize SARPES performance, we measured reflectivities and Sherman functions for Bi(111) at scattering kinetic energies from 3.0–5.5 eV. Total photoelectron intensity  $I_0$  without scattering was measured by detector 1 behind Lens 1 with the target removed. Scattered photoelectron intensity  $I$  was measured by detector 2 behind Lens 2. Reflectivity  $R = I/I_0$ , and single-channel efficiency is  $FOM = S^2$ , where  $S$  is the Sherman function. Figures 4(e) and (f) show reflectivities, Sherman functions, and FOMs at different energies. Error bars for Sherman function ( $\sigma_S$ ) and reflectivity ( $\sigma_R$ ) represent statistical errors from 4 and 100 measurements, respectively. FOM error bars are calculated via error propagation:  $\sigma\{FOM\} = \sqrt{(2\sigma_S S)^2 + (\sigma_{-S^2})^2}$ . Reflectivity increases with scattering kinetic energy, while Sherman function generally decreases. FOM initially increases then decreases, reaching an optimal value of  $2.1 \times 10^{-2}$  at 3.5 eV scattering kinetic energy.

Energy resolution was evaluated by measuring the Fermi edge width of the energy distribution curve (EDC) for a polycrystalline Au film grown by molecular beam epitaxy, using both 90 eV synchrotron radiation and 8.43 eV Xe lamp photons at 17.5 K. Energy-angle (E-A) images from ARPES and SARPES modes with 90 eV synchrotron radiation are shown in Figs. 6(a) and (e), acquired with 5 eV analyzer pass energy and 0.2 mm entrance slit width. Instrumental energy resolutions were determined by fitting Fermi edges (Figs. 6(b) and (f)) using a Gaussian function convolved with a linear background and the Fermi-Dirac function at sample temperature. The Gaussian full width at half maximum (FWHM) directly measures instrumental resolution, yielding 4.0 meV for ARPES and 5.3 meV for SARPES.

Angular resolution was measured using an angular device consisting of a mesh and 0.1 mm diameter gold-plated tungsten wire separated by 25 mm. Photoelectrons emitted at different angles from the wire pass through the mesh, creating a light-dark pattern shown in Figs. 6(c) and (g) for ARPES and SARPES modes. Angular resolution characterizes the system's ability to distinguish photoelectrons with similar emission angles. After passing through a mesh, the ideal angular intensity distribution would be a step function, but finite resolution broadens this distribution. The first derivative can be fitted with a Gaussian function whose FWHM characterizes angular resolution. Using 90 eV synchrotron radiation with 5 eV pass energy, angular resolutions are  $0.19^\circ$  and  $0.27^\circ$  for ARPES and SARPES modes, respectively (Figs. 6(d) and (h)). Using the same method with Xe lamp photons and 2 eV pass energy, energy resolutions are 6.2 meV (ARPES, 0.5 mm slit) and 13 meV (SARPES, 0.8 mm slit), while angular resolutions are  $0.32^\circ$  and  $0.29^\circ$ .

Table 1 summarizes all energy and angular resolution results for both ARPES and SARPES modes.

**TABLE 1.** Energy and angular resolutions of the spin spectrometer in ARPES and SARPES modes.

Condition	Energy Resolution	Angular Resolution
90 eV (ARPES)	4 meV	$0.19^\circ$
90 eV (SARPES)	5.3 meV	$0.27^\circ$
8.43 eV (ARPES)	6.2 meV	$0.32^\circ$
8.43 eV (SARPES)	13 meV	$0.29^\circ$

The spin polarimeter entrance aperture measures  $15 \times 12 \text{ mm}^2$ , corresponding to an energy window of 160 meV at 5 eV pass energy and an angular window of  $12^\circ$ . This yields approximately  $30 \times 44 = 1320$  distinguishable channels for energy and angular resolutions of 5.3 meV and  $0.27^\circ$  in SARPES mode. Considering the number of distinguishable channels  $N$ , total spectrometer efficiency is  $\text{FOM}_{\text{total}} = N \times \text{FOM} = 28$  at 3.5 eV scattering kinetic energy.

Table 2 compares the performance of representative SARPES stations and spin polarimeters worldwide, including energy resolution, angular resolution, single-channel efficiency, and multi-channel capability.

**TABLE 2.** Best performance of various SARPES stations and related spin polarimeters.

System	Energy Resolution (meV)	Angular Resolution (°)	Efficiency	Multi-channel	Source type
VLEED8 [?] Spin-LEED [?]		1.7 [?]	$1.9 \times 10^{-4}$ [?]	Yes	Xe light (8.43 eV)
180° VLEED [?]	0.2 [?]	0.7 [?]	$1.1 \times 10^{-2}$ [?]	Yes	Laser (6.994 eV)
This work	5.3	0.27	$2.1 \times 10^{-2}$	Yes	Xe light/Synchrotron Radiation

Our spin polarimeter is the world's only multi-channel system equipped with a synchrotron radiation source. Regarding measurable spin polarization components, Mott and VLEED spin polarimeters measure two components, while spin-LEED polarimeters based on 90-degree reflection measure only the component perpendicular to the scattering plane (two components maximum even with dual arms). In contrast, the 90-degree reflection VLEED polarimeter can potentially measure all three spin polarization components through target rotation and addition of a second reflection arm.

#### IV. Summary

We have developed a new image-type VLEED spin polarimeter employing 90-degree reflection at the BL07U beamline of SSRF. By measuring iron target reflectivities and SARPES asymmetries for Bi(111) at various scattering kinetic energies, we achieved an optimal single-channel efficiency of  $2.1 \times 10^{-2}$  at 3.5 eV scattering kinetic energy. The SARPES energy and angular resolutions reach 5.3 meV and  $0.27^\circ$ , respectively, with 1320 distinguishable channels and total polarimeter efficiency of 28. Furthermore, the 90-degree reflection VLEED design enables measurement of all three spin polarization components, with plans to add a second reflection arm in future BL07U beamline upgrades.

**Author contributions:** Mingzhu Zhang: Established and optimized the SARPES system, acquired and analyzed data, developed control software,

and prepared the manuscript draft. Muyang Wu: Designed, installed, and commissioned the low-temperature sample manipulator. Bo Zhao: Established and optimized the SARPES system, provided supervision. Shan Qiao: Established and optimized the SARPES system, acquired and analyzed data, secured funding, administered the project, provided supervision, and revised the manuscript.

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*Note: Figure translations are in progress. See original paper for figures.*

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