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Performance of a Mott detector for undulator-based spin-resolved spectroscopy

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To measure spin-polarized core-level electron spectra, a spectrometer equipped with a highly efficient retarding-potential Mott spin polarimeter using undulator-based soft-x-ray beamlines has been set up. With a thin film of Au as a target this polarimeter has an efficiency estimated to be $\sim 2 \times 10^{-4}$. The performance of this system for spin-polarized spectroscopy has been tested using core-level spin-polarized photoemission of magnetic and nonmagnetic thin films excited with linearly and circularly polarized light, respectively. Measurements using a new spin-resolved absorption technique are also discussed. © 2002 American Institute of Physics.

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I. INTRODUCTION

Photoemission and soft x-ray absorption are powerful experimental techniques for probing the electronic structure of matter. An important extension is spin-polarized electron spectroscopy.^{1,2} In addition to physical quantities such as kinetic energy and momentum measured in conventional photoemission spectroscopy, the measurement of electron spin polarization is also included in a spin-polarized spectroscopy. As an example, spin-polarized photoemission spectroscopy with linearly or circularly polarized incident photons has successfully revealed the spin-dependent electronic structure of a range of materials.³ The projection of spin polarization P along a given direction, e.g., the x axis, is defined as $P_x = (N_{\uparrow} - N_{\downarrow}) / (N_{\uparrow} + N_{\downarrow})$, where N_{\uparrow} and N_{\downarrow} are the numbers of electrons with spin projected onto the x axis being parallel and antiparallel to the positive direction of the x axis, respectively. The measurement of spin polarization, however, is not an easy task due to the low efficiency of available electron spin polarization analyzers.

In the past decade, considerable progress has been achieved in the technology of electron polarimetry and soft x-ray beamline. Spin polarimeters of several types have been developed based on the use of Mott scattering,⁴⁻¹³ diffuse scattering,¹⁴⁻¹⁷ low energy electron diffraction¹⁸ and spin-dependent scattering from magnetic thin films. Of these, Mott polarimeters are the most extensively used, but their efficiency is rather small, generally between 2×10^{-5} and 1.6×10^{-4} . Further improvement is required.

In this article we describe the design of a high-efficiency retarding potential Mott spin polarimeter for spin-polarized core-level electron spectroscopy used at undulator-based soft-x-ray beamlines. The efficiency of this polarimeter is estimated to be $\sim 2 \times 10^{-4}$ when a Au film is used as the scattering target. The performance of this spin-polarized electron spectrometer system has been successfully tested using core-level photoemission from nonmagnetic materials excited by circularly polarized light, and spin-polarized core level and valence band photoemission from magnetic thin films and spin-resolved x-ray absorption of magnetic oxides. This spin polarimeter can be extended to a spin polarization analyzer that provides *in situ* analysis of all three components of the spin polarization vector of electrons.

II. SPECTROMETER WITH SPIN ANALYSIS

A. Spectrometer

A new end station for spin polarized core-level electron spectroscopy has been established for use at the elliptically polarized undulator (EPU)¹⁹ and the U5 beamline of the Synchrotron Radiation Research Center (SRRC) in Taiwan. The EPU can generate circularly polarized light or linearly polarized light with the polarization in the horizontal or vertical direction with respect to the storage ring.²⁰ This end station is equipped with cryogenic sample cooling, sputtering, LEED, sample load-lock, and film growth capabilities, and is mainly used to explore the electronic structure of magnetic

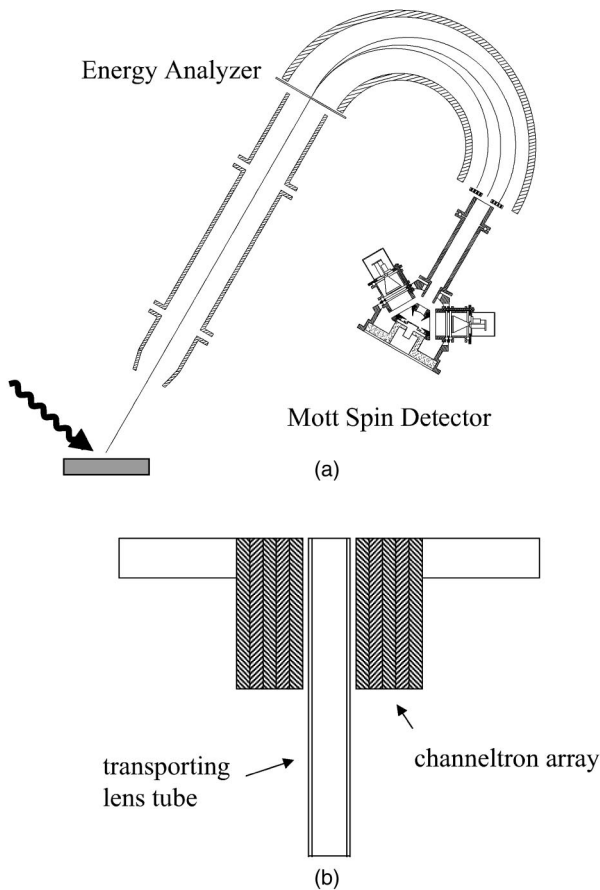


FIG. 1. (a) Overview of the spin-resolved spectrometer system. An electron multiplier array assembly located in the exit plane of the energy analyzer is used for non-spin-resolved multichannel detection. (b) Detailed cross-sectional view of the electron multiplier array assembly. Five 2×10 -mm-rectangular electron multipliers are on both sides of the central tube which transports the electrons to the Mott detector.

materials. The spectrometer is a VSW HA150 hemispherical electron energy analyzer to which a Mott spin polarimeter is adapted, as shown in Fig. 1(a). In the exit plane of the energy analyzer, a specially designed electron multiplier array assembly is used for non-spin-resolved multichannel detection. A rectangular aperture and a set of electrostatic cylindrical tube lenses connected to the electron multiplier assembly have been used to extract the electrons for spin-resolved measurements. Figure 1(b) presents the detailed cross-sectional view of the electron multiplier array assembly.

B. Spin polarimeter

In the Mott scattering of high energy spin-polarized electrons, the spin-orbit interaction results in a spatial asymmetry. The scattering cross section of electrons with spin polarization \mathbf{P} is described by the following equation:

$$\sigma(\theta) = I(\theta)[1 + S(\theta)\mathbf{P} \cdot \mathbf{n}], \quad (1)$$

where θ , $I(\theta)$, and \mathbf{n} are the scattering angle, the intensity of the scattered electrons without influence of the spin-orbit interaction and the unit vector perpendicular to the scattering plane, respectively; $S(\theta)$ is the Sherman function which is determined by the atomic number Z of the scattering target,

scattering angle θ and scattering energy. An essential feature of Mott scattering is that only the spin polarization vector perpendicular to the scattering plane contributes to the scattering asymmetry. In a Mott spin detector, a small instrumental asymmetry such as a difference in detector efficiency amounting to a few percent smears the polarization measurement. One method well established to eliminate the instrumental asymmetry is to reverse the spin polarization of the incident electrons. If the scattered intensities into equivalent left- and right-hand side solid angles are I_L and I_R , respectively, for incident electrons with spin polarization \mathbf{P} , and equivalently I'_L and I'_R for incident electrons with spin polarization $-\mathbf{P}$, the spin polarization perpendicular to the scattering plane can be deduced from

$$P = \frac{1}{S} \frac{1 - \alpha}{1 + \alpha}, \quad (2)$$

where α is defined as $\sqrt{I'_L \cdot I_R / I_L \cdot I'_R}$. One important parameter generally used to characterize the performance of Mott polarimeters is the efficiency ϵ , defined as

$$\epsilon = S^2 \frac{I}{I_0}, \quad (3)$$

where I_0 is the incident intensity, and I is the total scattered intensity given by $I = I_L + I_R$. Electron spin polarization can therefore be obtained from a measurement of the scattering asymmetry combined with the calibrated Sherman function of the polarimeter.

Mott polarimeters of various types employing a wide range of geometry and operation energy have been designed and constructed. An instrument of this type includes conventional high-energy Mott polarimeters^{4,6,11,12} and retarding-potential Mott polarimeters^{5,7,9,10,13} in which the incident electrons are accelerated by an electric field established between an inner and an outer-electrode. Although its Sherman function is larger than that of a retarding-potential Mott polarimeter, the drawbacks of a conventional Mott polarimeter are that the electron detector must be operated at high potential and that its discrimination against inelastically scattered electrons is poor. The compact conical-type retarding Mott spin polarimeter, or micro-Mott polarimeter, is widely used and exhibits the following features:⁹ (1) it is easy to operate, (2) it has a large electron acceptance phase space, and (3) it has a stable Sherman function. It is therefore easy to adapt a micro-Mott polarimeter to an energy analyzer for spin-polarized photoemission or Auger electron spectroscopy.

Several improvements of the micro-Mott polarimeter have been reported since its first development. These changes include the extension of its measurement from one transverse component to two orthogonal transverse components and the improvement of its efficiency. Defined earlier, the efficiency of an electron spin polarimeter is conventionally referred to as the figure of merit of a spin polarimeter

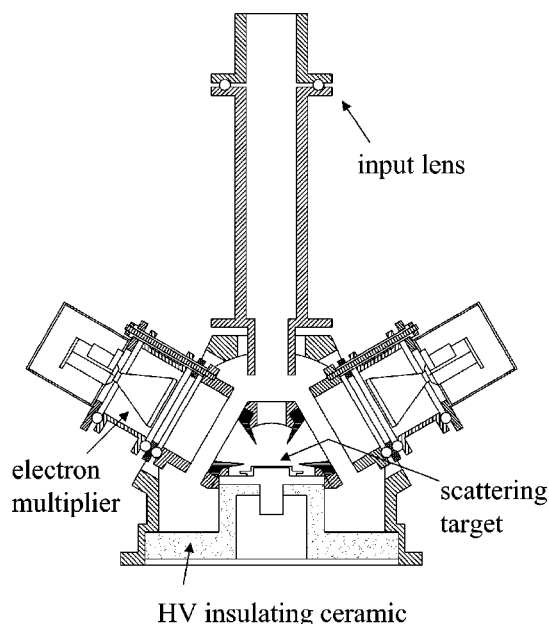


FIG. 2. Cross-sectional view of the Mott spin polarimeter with enlarged conic-type collection angle. Only one pair of electron multipliers is shown; the other pair is located in the perpendicular plane.

since it is proportional to the inverse square of the statistical error in an electron counting measurement that determines the spin polarization of the incident beam of electrons. The efficiency of a retarding-potential polarimeter tends to be low, but can be improved by increasing the effective Sherman function or the intensity of scattered electrons. Improvement of the Sherman function can be achieved by using high- Z materials, such as thorium or passivated uranium, as the polarimeter scattering target. With a bulk thorium target, nearly a factor of two gain in the figure of merit has been demonstrated.¹⁰ A further improvement, $S_{\text{eff}} \sim 0.20$ at a scattering energy of 30 keV, has been obtained by using a passivated uranium target. Another way of increasing the figure of merit is adequately choosing the energy loss window and enlarging the collection solid angle for detection of the scattered electrons.⁹

As calculated with a relativistic Hartree–Fock potential, the Sherman function and the differential cross section for Au are rather insensitive to the scattering angle in most Mott polarimeters with a mean scattering angle 120° .⁹ The scattering intensity integrated over the collection solid angle increases dramatically with increase of the collection solid angle at the expense of only a small degradation in average Sherman function. Because the figure of merit scales almost linearly with the solid angle of collection, the polarimeter efficiency can hence be improved on enlarging the solid angle collection.

Figure 2 displays the cross-sectional view of the design of the electron optics in the present spin polarimeter. Similar to previous micro-Mott polarimeters, this new polarimeter is a retarding potential polarimeter with a large (50°) conic type collection angle, i.e., a solid angle of 0.6 sr. The scattered electrons are decelerated by the electric field established between the outer-electrode of the polarimeter and the first focus electron lens in the collection optics. The electron

multipliers in the polarimeter operate near ground potential. In order to diminish the complexity in the collection lens optics, the electron multipliers are with a diameter: 25 mm, as shown in Fig. 2. The scattering target of the Mott circular opening spin polarimeter used in the performance test described in the following is an $\sim 100 \mu\text{m}$ Au thin film, and the scattering energy and the inelastic energy-loss window are 23 keV and 700 eV, respectively. Based on the results of the performance test, the effective Sherman function and the efficiency of the new polarimeter are estimated to be 0.13 and $\sim 2 \times 10^{-4}$, respectively. The efficiency can be improved to $4\text{--}5 \times 10^{-4}$ with Th or U as the scattering target.

III. PERFORMANCE TEST

The performance of the spin detector has been tested by performing spin-polarized $3s$ core-level photoemission from Fe thin films epitaxially grown on a W(110) substrate, spin-polarized photoemission of Pt $4f$ core-level photoemission excited by circularly polarized light, and spin-resolved absorption of magnetic oxides. An identical polarimeter has also been connected to the exit plane of a Scienta SES 200 analyzer and used for valence band studies of ferromagnetic gadolinium.²²

A. Spin-polarized Fe $3s$ core-level photoemission

The electronic properties of magnetic thin films have received great attention in both fundamental research and technological applications. Spin polarized photoemission has great potential for providing direct information on the magnetic and electronic structure of these materials. Some multiplets of photoemission from transition metals reflect the exchange interaction between the valence electrons and the unpaired core electrons left after the photoexcitation. The $3s$ photoelectrons from transition metals, for instance, are split into two multiplets that are not present in studies of nonmagnetic material systems.²³ A simple atomic model due to Van Vleck²⁴ has been used to interpret this splitting as the energy difference between a high spin and a low spin multiplets and, as such, the splitting has been considered as a finger print of the local valence spin moment. Recent spin polarized photoemission studies of transition metals qualitatively support this simple argument.^{26,27}

We have measured spin-resolved $3s$ core-level photoemission spectra of Fe thin films. Thin Fe films were epitaxially grown on W(110) substrates. Linearly polarized synchrotron radiation with an energy of 265 eV incident at an angle of 60° was used to excite photoelectrons which were detected in the direction normal to the sample surface. The Fe films were magnetized along [110] by applying current pulses through a pair of Helmholtz coils. The current pulse was generated by means of discharging a bank of capacitors through four high power SCRs connected as a H bridge in either direction.²⁵ The spin polarization was measured in the remnant state of the films and the spin polarization induced in the Mott detector resulted from spin-orbit effect was eliminated by averaging two sets of data with opposite magnetization. In agreement with earlier studies, our results, Fig. 3, clearly show that the binding energy of the high-spin final

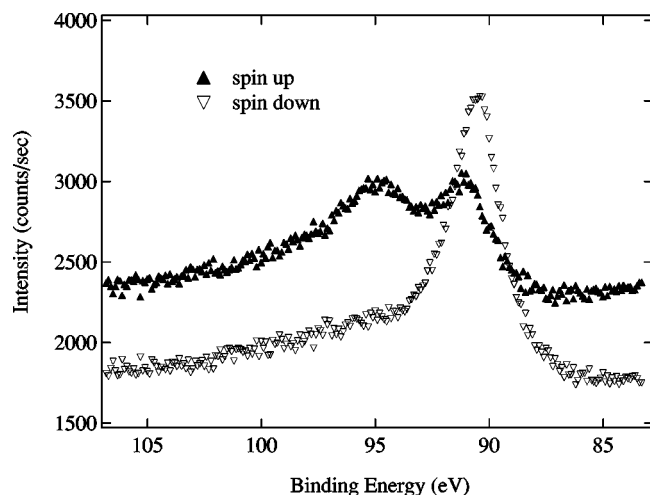


FIG. 3. Spin-resolved $3s$ photoemission of Fe thin films epitaxially grown on W(110) substrates. Linearly polarized synchrotron radiation with an energy 265 eV was used to excite the photoelectrons. The total energy resolution was 0.35 eV.

state is lower than that of the low-spin final state. Furthermore, the high-spin final state is composed of both majority spin photoelectrons with a binding energy 91.0 eV and minority spin photoelectrons with a binding energy 90.4 eV, but only majority spin exists in the low-spin final state with a binding energy 95.0 eV. Interestingly, the present study again shows that the two spin components in the high-spin state are separated in binding energy as reported previously.²⁷ It is important to note for the purposes of the present study that the increased efficiency of the present detector results in an improved signal-noise ratio when compared to the earlier studies.^{26,27}

B. Core-level photoemission with circularly polarized light

Spin-polarized photoelectrons can be produced from unpolarized atoms with circularly polarized, linearly polarized, or even unpolarized light due to the “Fano Effect.” The spin-orbit interaction is the essential mechanism for the latter effect and the spin polarization is induced by an interference between the dipole allowed final state wave functions with angular momentum $l+1$ and $l-1$ in the presence of spin-orbit interaction.²⁸ This effect has been experimentally observed in alkali-metals, rare-gas atoms, and semiconductors such as GaAs, a widely used material for spin polarized sources. Photoelectrons excited from the $4f$ core level of W(110) by circularly polarized light have been observed to be highly spin polarized, in agreement with free atom theory.²⁹ Such a system is, therefore, very suitable for testing a spin polarimeter on a synchrotron beamline. We carried out such a measurement on the $4f$ core level of Pt(111) excited by 300 eV circularly polarized light. As shown in Fig. 4, our results reveal that Pt $4f$ photoelectrons are highly spin polarized with opposite spin polarization in $4f_{7/2}$ and $4f_{5/2}$ states. This test demonstrates both the high quality of circularly polarized light produced from the EPU beamline and good performance of the Mott spin detector.

As demonstrated in the measurement of spin singlet states in high temperature superconductor mother

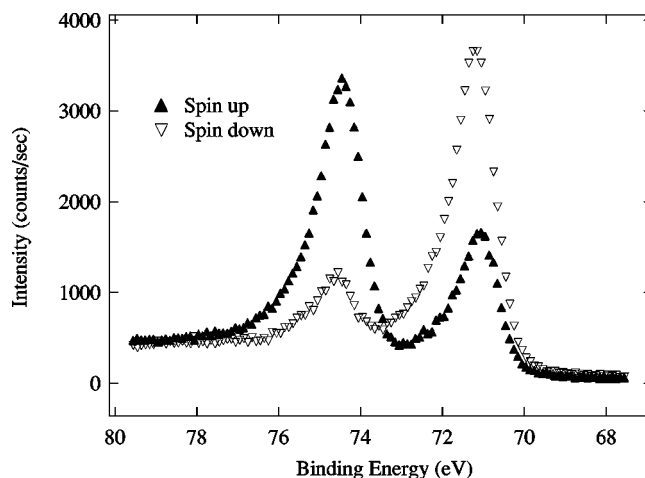


FIG. 4. Spin-resolved Pt $4f$ photoemission excited circularly polarized from the EPU beamline. The degree of the light circular polarization is better than 90%. The total energy resolution was 0.35 eV.

compounds,^{30,31} spin polarized photoemission using circularly polarized light provides useful information of the spin character of antiferromagnetic materials. With this type of photoemission spectra, quantitative information on spin and orbital moments in macroscopically nonferromagnetic materials, in particular transition-metal oxides, can also be obtained. The integration of the difference between the spin-up and the spin-down spectra excited by circularly polarized light yields $\langle L \cdot S \rangle$ like expectation values.³²

C. Spin-resolved O K -edge x-ray absorption

Soft x-ray absorption at the oxygen K edge is a dipole allowed transition in which an O $1s$ electron is excited to an empty O $2p$ orbital. In transition metal oxides, oxygen $2p$ states are hybridized with the $3d$ states of transition metals. Oxygen K edge x-ray absorption spectroscopy provides valuable information on the unoccupied electronic structure, and is closely related to inverse photoemission, another probe of the unoccupied bands. Soft x-ray absorption spectra can be obtained by measuring the sample drain current, the intensity of transmitted x-ray and Auger electron intensity of the core-hole decay. Several theoretical studies have shown that the spin triplet in the KLL Auger transition is forbidden, and only the spin-singlet is allowed.^{21,33} The two-hole final state in the KLL Auger transition of half metallic CrO_2 have been experimentally shown to be a spin singlet.³⁴ With the on and off O K -edge-resonant ($1s \rightarrow 2p$) excitation, the dramatic difference in the spin polarization shows that the exchange correlation effect prevents the two-hole Auger final state from being a spin-triplet state. This effect points to the possibility of an important application of spin-resolved x-ray absorption in magnetic oxides where the oxygen plays an important role in magnetism via superexchange or double exchange interactions. The spin selection rule provides a powerful method for spin-resolved O $1s$ absorption spectroscopy. With the spin selection rule, the spin-resolved O $1s$ absorption spectroscopy can be achieved by monitoring the spin resolved Auger electrons which are emitted at a constant kinetic energy. The spin character of the O $2p$ states above the Fermi level can be revealed by using spin-resolved O $1s$

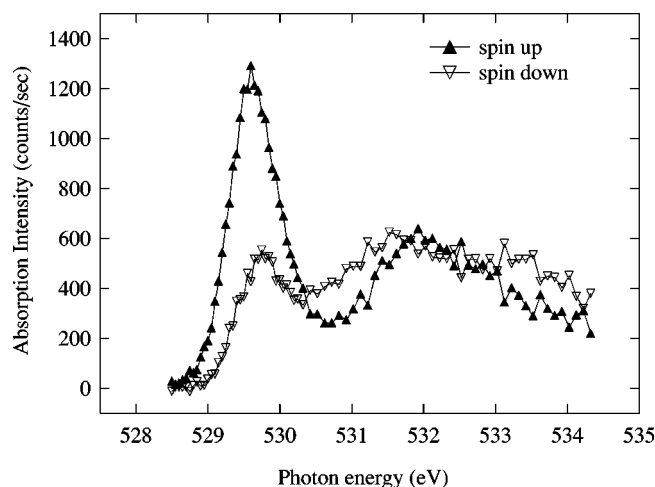


FIG. 5. Spin-resolved O $1s$ XAS spectra of CrO_2 , taken in the partial electron yield mode on monitoring the O KLL Auger electrons. The photon energy resolution and the electron analyzer resolution were 0.3 and 0.6 eV, respectively.

XAS. This technique is complementary to the MCD in O K -edge absorption, which results predominately from the O $2p$ -projected orbital magnetization.

We applied this new technique to explore the spin-polarized unoccupied states of CrO_2 . O $1s$ XAS spectra in a spin-resolved mode has been obtained by collecting the O KLL Auger partial electron yield. Figure 5 displays the spin-resolved O $1s$ XAS spectra of high-quality CrO_2 thin films grown on $\text{TiO}_2(100)$ which provides an ideal template. The photon energy resolution was set to 0.4 eV with \mathbf{E} perpendicular to the c axis. The measurement shows that the lowest energy states, i.e., the states closest to the Fermi level, have a spin-polarization of $(85 \pm 10)\%$. The states at the Fermi level are therefore almost fully spin-polarized, consistent with the predictions by band structure calculations³⁵ and the Andreev reflection measurements at the superconductor-metal interfaces.³⁶

Surprisingly however, the spin polarization of the prepeak at 529.6 eV is only 50%, while all band structure calculations predict that the polarization of the lowest major structure of the conduction band should have been 100%. This large discrepancy indicates that strong correlation effects are present in the system, since in such a case, the concept of density of states as defined in effective one-particle theories loses its meaning as a basis for a quantitative understanding of excitation spectra, i.e., density of states are then quite different from spectral weights. The spin-resolved O K -edge XAS spectra of CrO_2 provide valuable information on the unoccupied O $2p$ electronic states which are strongly hybridized with Cr $3d$ states. Detailed analysis of these data will be discussed elsewhere.

IV. FUTURE IMPROVEMENT: A SPIN POLARIZATION ANALYZER

In some spin-polarized electron spectroscopy experiments, *in situ* complete measurements of all the components of a spin polarization vector is valuable, for instance the studies of magnetic anisotropy in ultrathin films or surfaces.

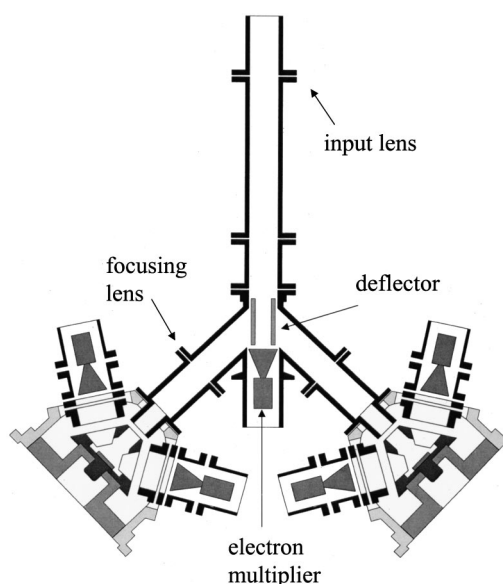


FIG. 6. Design of a future spin-polarization analyzer that incorporates two polarimeters and an electrostatic deflector. Only one pair of electron multipliers are shown in each polarimeter.

However only the transverse spin polarization components of incident electrons are measurable in a Mott spin polarimeter. The measurement of the longitudinal component can be achieved if the electron beams are deflected such that the transverse spin component is converted to the longitudinal component. Figure 6 illustrates the design of a spin-polarization analyzer which permits *in situ* analysis of all the three components of a spin-polarization vector.³⁷ This new spin polarization analyzer incorporates two polarimeters and an electrostatic deflector. The energetically selected electrons can be deflected $\pm 45^\circ$ either into the left or the right spin polarimeter by an electrostatic deflector for spin-polarized measurements. In addition, electrons passing straight through the deflector can also be focused onto an electron multiplier located at the center for non-spin-resolved measurements. Measurements of core-level spectra can be easily switched between, with, and without spin analysis. In both spin polarimeters, spin polarization normal to the scattering plane is measured via the spatial asymmetry in the Mott scattering. Four electron multipliers in each spin polarimeter detect the two orthogonal transverse spin-polarization components simultaneously. This new spin-polarization analyzer, therefore, can measure three orthogonal spin-polarization components *in situ*. Combined measurements of the two spin polarimeters determine the two orthogonal components in the deflection plane of the electrostatic deflector, and also provide a self-consistent check of component measurement along the direction perpendicular to the deflection plane.

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