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T. Komesu, G. D. Waddill, J. G. Tobin

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Takashi Komesu, G.D. Waddill,
University of Missouri-Rolla, Department of Physics, Rolla, MO 65401-0249
and J.G. Tobin
Lawrence Livermore National Laboratory, Livermore, CA 94550

Abstract:

Spin-polarized electron spectroscopies are used to probe the nature of magnetism in surfaces and thin films. Strong inelastic scattering of the electron from particle-hole and collective excitations results in short mean free paths that can be spin dependent. A quantitative understanding of the spin dependence of the inelastic mean free path is critical to the interpretation of results from spin-polarized electron spectroscopies such as spin-polarized low energy electron diffraction, spin-polarized electron energy loss spectroscopy, and spin-polarized photoemission. Previous studies measured such effects for Cu 3d electrons that were photoexcited with sufficient energy to pass through Fe [1] and Co films [2] grown on the Cu. In these studies, the kinetic energy of the Cu 3d photoelectrons was typically less than 50 eV. Substantial spin asymmetry of the photocurrent was observed and interpreted as being due to the spin-dependence of the photoelectron inelastic mean free path. In this paper, we have examined such spin dependent behavior of electrons transmitted though magnetic films with higher kinetic energy in order to reduce complications due to spin-flip scattering which are more prevalent at lower kinetic energy [1, 2]. As in the earlier studies [1, 2] we observe clear evidence of a spin filter effect for the Ag 3d photoelectrons through a ferromagnetic Fe film.

Introduction

Spin-polarized electron spectroscopies such as spin-polarized electron energy loss spectroscopy and spin-polarized photoemission require knowledge of the spin-dependent transmission of the electron through magnetic materials in order that a proper assessment of the experimental data can be achieved. Experimental [1-3] and theoretical [4, 5] studies have reported on the spin-dependent transmission of electrons through thin magnetic films. The results show that more majority spin electrons are transmitted through magnetic films than minority spin electrons, but the interpretation of this experimental result, observed for ferromagnetic films grown on Cu(001) [1, 2], is still the subject of some controversy. Also, prior to these photoemission-based transmission measurements, the spin polarization of secondary electrons from Ni(110) was observed to be influenced by spin-dependent inelastic scattering [6] that was modeled in a recent paper [7]. In addition, spin-dependent electron lifetimes have been observed with majority spin electrons having much longer lifetimes than minority spin electrons [8]. All these results were initially explained in terms of a spin-dependent inelastic mean free path for the electrons, but some theoretical studies showed that the results could be understood in terms of spin-dependent *elastic* scattering processes and that inelastic processes were not of sufficient strength to explain the experimental data [4, 5]. However, the direct electron transmission measurement using spin-polarized electrons [9] shows significant polarization values not only for elastically scattered electrons, but also for inelastically scattered electrons. In all the above-mentioned experimental and theoretical studies, electrons with kinetic energies below ~50 eV were used. This complicates the interpretation because in this energy region there can be significant contributions to the transmission due to spin-flip exchange scattering [10], and spin-dependent band structure [11]. Because earlier studies concentrated on valence band photoemission [1-3], the substrate signal had to be isolated from the photoemission signal from the ferromagnetic film by subtraction of the ferromagnetic film contribution. However, the spin dependent band structure is sufficiently complex that this background subtraction procedure may not be completely reliable. For these reasons we have reexamined electron transmission through magnetic thin films for higher kinetic energies (720 eV for $Ag3d_{5/2}$ photoexcited electrons) where contributions from spin-flip exchange scattering should be greatly reduced [10].

The spin scattering asymmetry responsible for the spin filtering effect seen in the spin dependent mean free path studies has at its origin the spin imbalance between majority and minority spin electrons in the ferromagnetic thin film [8]. With fewer empty majority spin states a longer lifetime for majority spin electrons is expected relative to minority spin electrons, and minority spin electrons are filtered out upon passage through a ferromagnetic film. Consequently, further studies extending spin-dependent mean free path data should contribute to a better understanding of spin-dependent electron transport through magnetic materials.

Experiment

The main components of the spin-resolved photoemission spectroscopic experiment are described in reference [12], and a simple diagram is shown in Fig. 1. The photoexcited electrons are collected by a hemispherical electron energy analyzer (Physical Electronics Model 3057) with multichannel electron detection. For spin-resolved work, the multichannel detector has a central hole that allows passage of the energy analyzed electrons into the electron optics for the mini-Mott detector. In this case, the high voltages on the channelplates are turned off and the channelplates and anode assembly become part of the first lens stack, directing the electrons into a 90° spherical sector. The 90° sector is run at relatively high pass energy: the ultimate energy resolution is determined by the hemispherical analyzer pass energy and the photon source. After the 90° sector, the electrons travel through another lens stack, into the mini-Mott detector. In the mini-Mott, the electrons are accelerated to ~25 kV, with four channeltrons positioned horizontally and vertically used for electron counting. This allows the simultaneous resolution of spin-polarization both normal to the sample and in the plane of the sample perpendicular to the plane containing the emission direction and the sample normal. For the measurements here, the incident photon angle was 55° from the sample normal, and electrons emitted normal to the sample were detected. The overall energy resolution for the core level measurements (photon and energy analyzer) is about 0.7 eV full width half maximum or better. The experiments were carried out at 1×10^{-9} Torr or lower, and all measurements were at room temperature using a photon energy of 1100 eV.

The Fe/Ag(001) system has been studied by numerous groups [13-16] and offers a number of advantages in the current study. While there is still some controversy over details of the Fe/Ag(001) system what is established is that at low coverages the Fe easy axis is perpendicular to the film at room temperature while for thicker films (> ~7 ML) the easy axis is in the plane of the Fe film. The structure of the Fe films in this higher coverage regime has been identified as either the bcc structure of α-Fe or a slightly tetragonally distorted variant of the bcc structure. All films examined in this study were of the thicker variety with an in-plane easy axis of magnetization. For the spin-dependent transmission measurements, we monitored the spin-polarization of the Ag $3d_{5/2}$ core level emission (368.5 eV binding energy). This has the advantage of being well separated energetically from any Fe core level peaks, and it is therefore not necessary to remove any background polarization that might arise from Fe features or from Fe derived polarization in the secondary electron background near the Ag $3d_{5/2}$ peak. This is in contrast with earlier spin-dependent transmission measurements at lower energies that required subtraction of the spin-polarized Fe and Co valence bands to extract the spin-polarized Cu substrate signals.

The Ag crystal was cleaned with Ar^+ sputtering followed by annealing to about 450 °C and shows no evidence of contamination in the XPS scans. Fe films were deposited using e-beam evaporation. The films have a p(1x1) LEED pattern, are epitaxial to the Ag(100) substrate, and show less than 1% of O_2 contamination as judged from XPS measurements. The film thickness was determined from the intensity ratio of the Fe 2p (binding energy, $2p_{1/2}$ = 720 eV and $2p_{3/2}$ = 707 eV) to the Ag 3d (binding energy, $3d_{3/2}$ = 374.4 eV and $3d_{5/2}$ = 368.5 eV) peaks. Here the intensity ratio may be written as

$$\frac{I_{Fe}}{I_{Ag}} = \frac{I_{Fe}^0}{I_{Ag}^0}\left(\frac{1-e^{-x/\lambda_{Fe}}}{e^{-x/\lambda_{Ag}}}\right)$$

Where I_{Fe}^0 is the intensity of the Fe peak measured from a very thick film, and I_{Ag}^0 is the Ag peak intensity for a clean Ag film. The magnetization of the Fe films was done with an *in situ* pulse coil capable of magnetizing the samples either in plane or perpendicular to the plane of the films. All Fe films we examined were magnetized in the plane of the film (parallel to the [001] direction), and this was confirmed using x-ray magnetic circular dichroism (XMCD) acquired

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using beamline 4ID-C at the Advanced Photon Source at Argonne National Laboratory [17]. In addition, spin-polarized secondary electron spectroscopy with x-ray excitation was performed to check the Fe films magnetization. The secondary electron spin-polarization was measured using the mini-Mott detector described above for kinetic energies of ~ 3 eV, and this polarization is known to be related to the sample magnetization [18].

Results and Discussion

Figure 2 shows the spin-resolved Ag $3d_{5/2}$ core level spectra for a) clean Ag(001), b) a 19 ML Fe film on Ag(001) that was magnetized, and c) a 19 ML Fe film on Ag(001) that was not magnetized. In all cases, the majority electrons are indicated with upward pointing triangles and the minority electrons with downward pointing triangles. For the magnetized samples, an *in situ* pulse coil was used to magnetize the films parallel and antiparallel to the [001] direction and this was used to remove any instrumental asymmetry from the measurements [19]. For the clean Ag (Fig. 2a), the spectra show no spin-polarization as expected. For the magnetized 19 ML Fe film we clearly observe spin-polarization in the Ag $3d_{5/2}$ peak with greater intensity for the majority spin electrons and a net polarization of approximately 5% at the peak. Not shown here are results obtained for thinner Fe films (but still sufficiently thick to have a magnetic easy axis in the plane of the film). For these films we also observed a majority spin polarization, but with a reduced polarization. These results are consistent with the longer mean free path of the electrons in these films at the higher kinetic energies we are measuring, and with the results of spin polarized secondary electrons from these films that will be discussed below. Finally, for the 19 ML Fe film that was NOT magnetized during the measurements we again see no spin polarization in the Ag 3d core level as expected. These results are consistent with previous reports for Fe/Cu [1] and Co/Cu [2] using lower kinetic energy photoelectrons that have been interpreted in terms of a longer inelastic mean free path for majority than for minority spin electrons in a ferromagnetic medium. For our measurements, it is critical that no spin polarization is observed on the low and high binding energy sides of the Ag 3d emission or in the region between the Ag $3d_{5/2}$ and $3d_{3/2}$ peaks that might be from an iron derived secondary electron background. Further confirmation of the validity of our results comes from the absence

of any observed spin polarization in the Ag photoemission upon traversing an unmagnetized Fe film (Fig. 2c). Comparing with the results for the ferromagnetic film in Fig. 2b demonstrates that there is no spin dependent transmission through a film with no net magnetization.

In Fig. 3, we summarize the Ag photoelectron polarization values measured after transmission through Fe films of various thicknesses. The square data points are for spin polarized secondary electron spectroscopy with x-ray excitation. Here we see a secondary spin polarization that increases rapidly from zero polarization for Fe films near 10 Å thick with the polarization of the films saturating for films near 15 Å in thickness at ~40%. This is very consistent with spin resolved inverse photoemission studies of the evolution of electronic and magnetic properties of Fe films on Ag(001) at room temperature where bulk behavior is achieved for films around 15 Å thick with thinner films exhibiting decreasing exchange splitting [13,14]. The circular data points in Fig. 3 are the averaged polarization of the Ag $3d_{5/2}$ core level peak as seen in Fig. 2. Again, we see no polarization for thin films, an abrupt increase in polarization near 10 Å with saturation achieved for thicker films (~20-25 Å). The dashed line is meant only to guide the eye. Comparing with the spin-polarized secondary results provides compelling evidence that the total polarization of Fe films and the transmitted electron polarization through ferromagnetic materials are correlated. The vertical dashed line at 9 ML (12.97 Å) is the inelastic mean free path from the NIST database [20].

Figure 4 shows the spin polarized attenuation behavior of the Ag $3d_{5/2}$ peak. As explained in detail in Ref. 2 it is possible to calculate the mean free path at a certain kinetic energy based on the attenuation of that photoemission feature with increasing Fe film thickness. Applying this method to our results gives an inelastic mean free path of about 16.4 Å (~11.4 ML) for majority spin electrons, and about 14.3 Å (~10 ML) for minority spin electrons.

Conclusion

We have carried spin-resolved photoemission spectroscopy (SRXPS) measurements on Ag 3d core level peaks to probe the spin dependent mean free path though ferromagnetic Fe films grown on the Ag(100) surface. As was observed in previous spin-dependent mean free path measurements for lower kinetic energy electrons, we see that majority spin electrons have a

longer mean free path than minority spin electrons. Furthermore, our results show that this spin-dependent mean free path persists to much higher kinetic energies where contributions to spin-dependent scattering in elastic channels should be negligible.

Acknowledgment

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[20]   NIST Standard Reference Database 71 NIST ELECTRON INELASTIC MEAN FREE PATH DATA BASE Version 1.1

Figure captions

Figure 1   Experimental setup for spin-resolved photoemission spectroscopy. Here photoexcited electrons from the Ag substrate pass through a thin ferromagnetic Fe film and are collected with a hemispherical energy analyzer. The photoelectron spin polarization is determined using a mini-Mott detector after energy analysis is done with the hemispherical analyzer.

Figure 2   Spin dependent Ag $3d_{5/2}$ core level spectra (hυ = 1100 eV) for various Fe overlayer conditions. The left panel a) shows the results for a clean Ag substrate with spin up (majority spin which is parallel to the applied external magnetic field) indicated by upward pointing triangles, and spin down (minority spin which is anti-parallel to the applied magnetic field) by downward pointing triangles. The center panel b) shows the results for transmission through a magnetized 19.2 ML Fe film on top of Ag(100). The right panel c) is the same as for b), but the Fe film has NOT been magnetized in this case.

Figure 3   Summarized results of the secondary electron spin-polarization of different thickness Fe films on Ag(100) (square symbols and left axis) that indicate the degree of magnetization of the films. The spin-resolved Ag $3d_{5/2}$ polarization through the same films is shown (circular symbols and right axis) for comparison. The similarity in the two curves suggests that the Ag core level spin polarization can be related to the net magnetization of the Fe films these electrons pass through. Here 1ML ≈ 1.8 Å for the bcc Fe structure.

Figure 4   Summarized log plot of the attenuation of the Ag $3d_{5/2}$ intensity as a function of Fe film thickness. The blue data points are for majority spin electrons and the red for minority spin electrons. The solid lines are the best fit to the data and are used to

extract the spin dependent mean free paths of 16.4 Å for majority spin and 14.3 Å for minority spin.
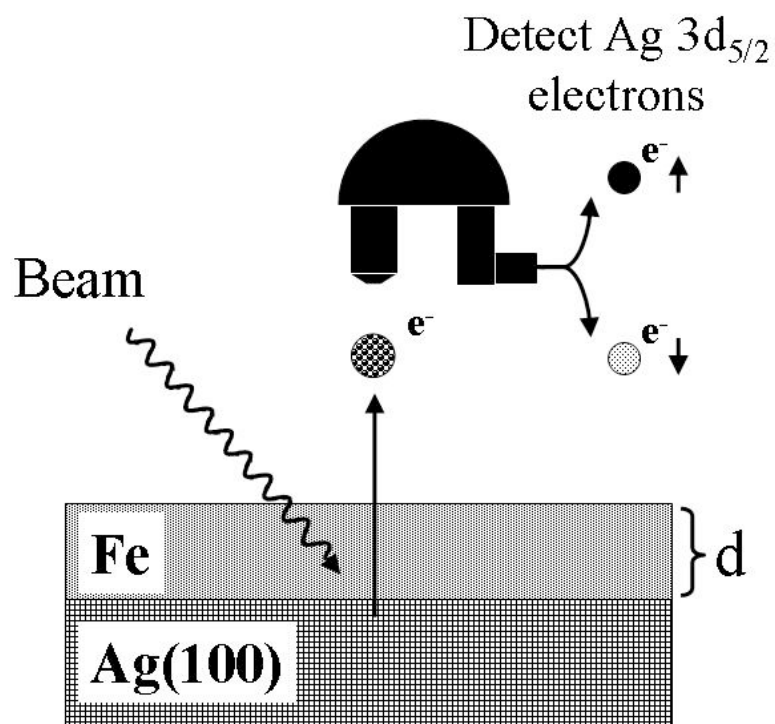
Detect Ag $3d_{5/2}$
electrons

e^- ↑

Beam

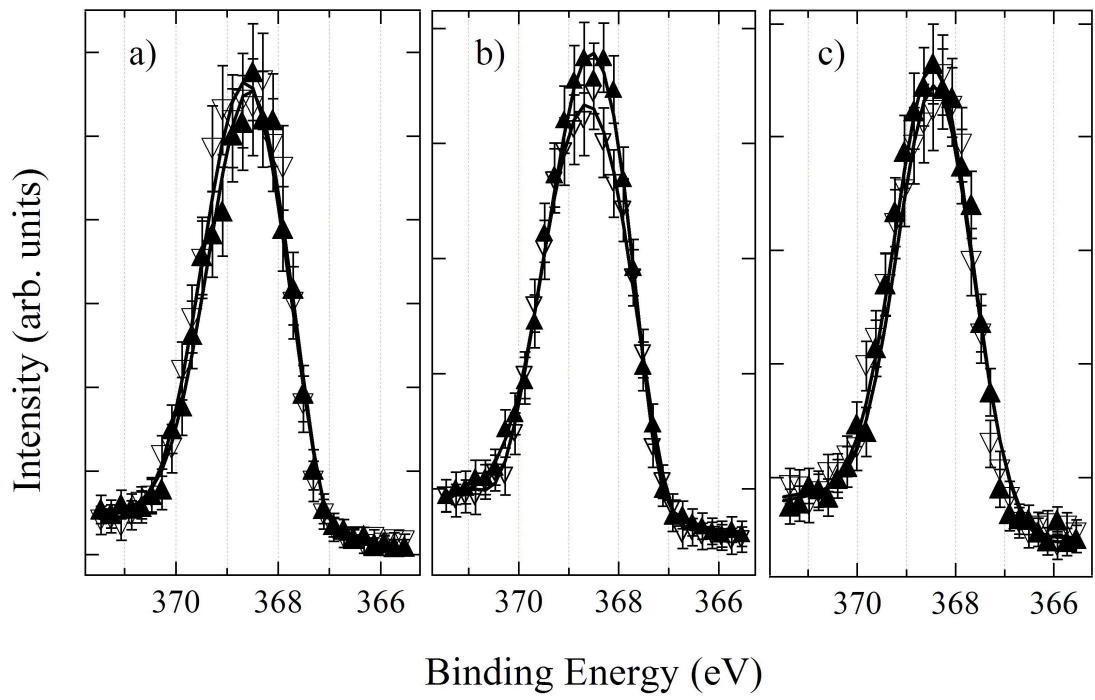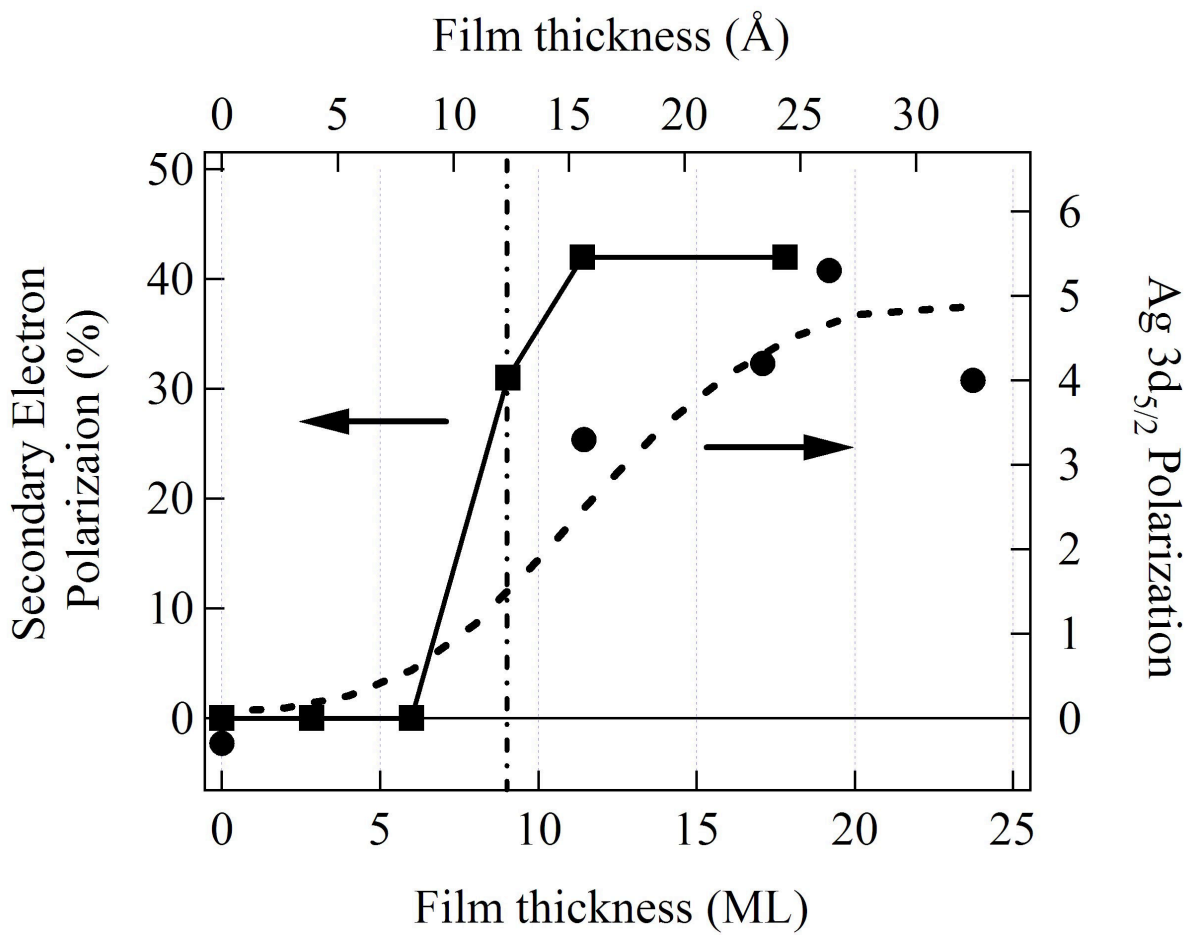e^-

e^- ↓

Fe

}d

Ag(100)

Figure 1, Komesu et. al
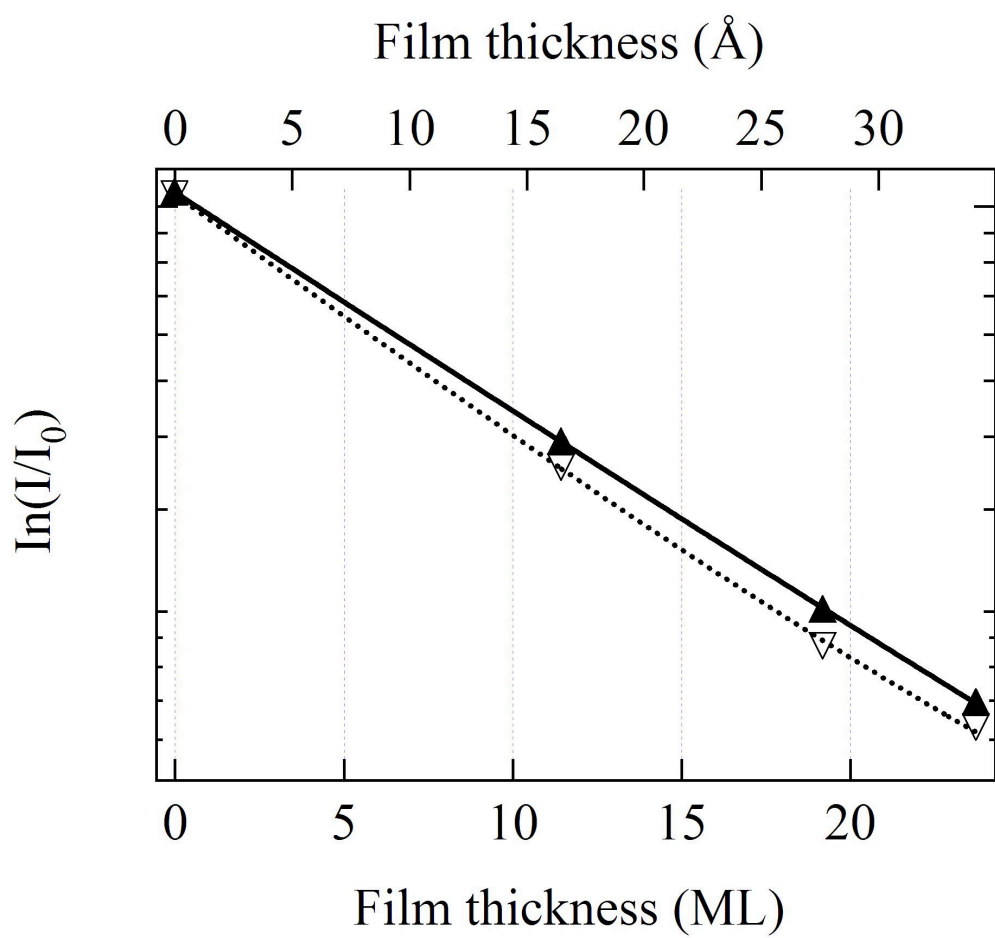
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Figure 2, Komesu et. al

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Figure 3, Komesu et. al

Figure 4, Komesu et. al