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Determination of the Photoabsorption Cross-Sections of Al and Fe Films in the Soft X-Ray Region Using Synchrotron Radiation *

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Abstract Photoabsorption cross-sections at the Al and Fe $L_{2,3}$ edges on thin films have been measured at the Beijing Synchrotron Radiation Facility (BSRF) using monochromatic soft X-ray radiation. The measured Fe and Al cross sections are in good agreement with theoretical predictions for energy below the edge, while a poor agreement is observed near the edge region probably because the independent atom approximation is not suitable for photon energy near the absorption edge. Experimental data have been also compared with previous data available in the literature. This work demonstrates that cross section determinations of thin films of low Z atoms in the transmission mode are difficult but are feasible and can be substantially improved using synchrotron radiation.

Key words photoabsorption cross-section, Al, Fe, soft X-ray, synchrotron radiation

1 Introduction

The photoabsorption cross-section is a fundamental physical quantity characterizing the interaction between photons and matter[1—15]. Accurate experimental data are important not only to improve the optical design of new instruments but also for evaluation of solid-state theoretical models. In the extreme ultraviolet and soft X-ray energy range (e.g., from 10eV to 1500eV), the photoabsorption cross-section of each material characterizes its optical properties and it is a basic quantity used to design optical coatings, multilayer reflectors and bandpass filters for many researches such as atomic physics, synchrotron radiation and X-ray astronomy. In addition, the comparison between theory and experimental photoabsorption cross-sections allows checking the quality of the wave-functions describing a material[1,4—6]. An accurate determination of the cross-section is also necessary to determine the continuum oscillator strengths of a theoretical model and to derive partial cross sections of a particular process.

In this manuscript we present data of the photoabsorption cross-section of freestanding films of Al and Fe for photon energies near the $L_{2,3}$ edge. Due to the relatively low absorption below the $L_{2,3}$ edge, these materials may be used as filters or as spacer in the manufacture of multilayer optics. Several studies of the absorption spectra of these two elements have been published[7—15], however, most of the previous investigations have been focused on the high energy cross section. In the soft X-ray energy region, the number of investigations of the photo-absorption cross section are limited mainly because of the experimental difficulties associated to the strong absorption of the materials in this spectral region. As a consequence of the high absorption only very thin samples may allows measurements, but these latter are very sensitive to surface contributions, e.g., presence of an overlayer due to, for example, a carbon contamination or an oxidation process.

For photon energies above 50eV the photoabsorption cross-section of an element may be determined using the tabulated atomic scattering factors in the framework of the independent atomic approximation, i.e., that atoms interact with radiation as if they were isolated. The approximation is valid for photon energies far from the absorption edge, but because the fine structures observed in a photoabsorption spectra near an absorption edge are determined by the local atomic environment[16], it is evident that near the edge the cross-section

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can not be a simply sum of the contributions of the individual atoms. Furthermore, the independent atom approximation is expected to breakdown at low energies (generally below 50eV) where the band structure may influence the optical properties of the materials.

In this manuscript we present and discuss photo absorption cross-section data of Al and Fe films performed using monochromatic radiation. The photon source we used is a synchrotron radiation source with the unique advantage to combine high flux over a continuous spectrum. Experimental results will be compared with data available in the literature.

2 Experiment

Measurements were performed at the Beijing Synchrotron Radiation Facility (BSRF) at the beamline 3W1B. Soft X-rays monochromatic radiation was obtained using a grating monochromator equipped with a plane grating with a variable line spacing characterized by a central line density of 1000 l/mm and a gold coated plane mirror. In our set up, with a fixed exit slit, energy is scanned rotating the grating and the mirror. In this energy range the best achievable spectral resolving power ($E/\Delta E$) is 300. The monochromator output is 10^{11} photons/s into a 0.1% bandwidth at 170eV. More details of the Beamline 3W1B are given in Ref. [17]. The beamline may tipically operate in two modes: parasitic (1.8GeV and max. current of 70 mA) or dedicated (2.2GeV with max. current of 100 mA). The measurement of the energy dependent cross-section in transmission mode are independent by the operation modes however, data presented have been collected during dedicated runs. During experiments the signal of the electron current circulating in the storage ring have been collected in real time to allow proper normalization of the decay of the electron current. In the experimental set up photons enter inside a reflectometer where both transmission and reflectivity of a sample can be recorded using a photodiode (AXUV-100, IRD, USA) and an electrometer (6517, Keithley, USA).

The samples used for our photo-absorption cross section measurements were Al and Fe films, manufactured by magnetron sputtering with 0.33Pa of Ar gas in a chamber with a base vacuum pressure of 10^{-4} Pa. The substrates were Si wafers coated with photoresist. Films were removed from the substrate by soaking in acetone. However, before the installation inside the vacuum chamber at a pressure of about 10^{-4} Pa, natural oxide layers were formed on both sides of the Al and Fe films. The thicknesses of these layers were estimated of about 0.1nm for Al and 0.5nm for Fe films respectively. Corrections for these oxide layers were considered as uncertainties of the data and in order to correct the photoabsorption cross sections an accurate determination of the film thickness is required. Thickness was measured on the freestanding films after they were removed from the substrate, measuring the energy loss of α particles passing through them. Both films were placed between an ^{241}Am α particle source and a Si surface barrier detector in a small vacuum chamber. The energy loss of 5.486MeV α particles was converted into thickness using the tables of the stopping powers of α particles[18]. Actually, this method determines only the mass per unit area, and returns a thickness for Al and Fe films of 0.513μm and 0.631μm respectively, using as bulk densities: 2.7g/cm^3 for Al and 7.8g/cm^3 for Fe. The relative uncertainty on the thickness is about 1% for Al and 0.7% for Fe. An alternative method[12] to measure film thickness with a better accuracy (about ten times better than the α particle method) have not be applied because of its complexity.

Transmission experiments were performed in the energy range 58—80eV with steps of 1eV for the Al film and in the range 410—1010eV with steps of 10eV for the Fe film.

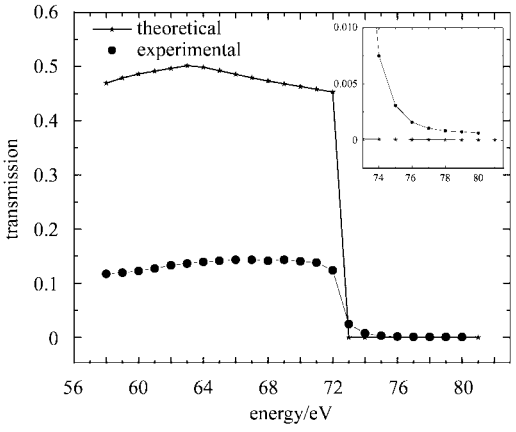

Fig.1. The measured transmission versus photon energy for a thin Al film.

The photoabsorption data for Al are shown in Fig.1. Curves report the Beer law $T = I/I_0$ e.g., the transmission T of the film, where I is the intensity of the beam transmitted through the sample and I_0 is the intensity of the incident beam. The most pronounced features of the transmission curves are the dip at the Al L_3 edge, but it is also evident

from Fig.1 that a great discrepancy exists between theory and experimental values below the absorption edge. We address this behavior to the enhanced absorption at the edge due to the surface film contamination and to the presence of the oxide layers. Above the edge, the present experimental results are larger than theory[19] (shown in the inset) because of the contribution of stray light and high-order harmonics present in the incident beam. So the measurements of cross sections is also affected by the existing high order radiation and stray light above absorption edge.

In Fig.2 we present the results of the transmission of the Fe film vs. the incident photon energy from 410eV to 1010eV.

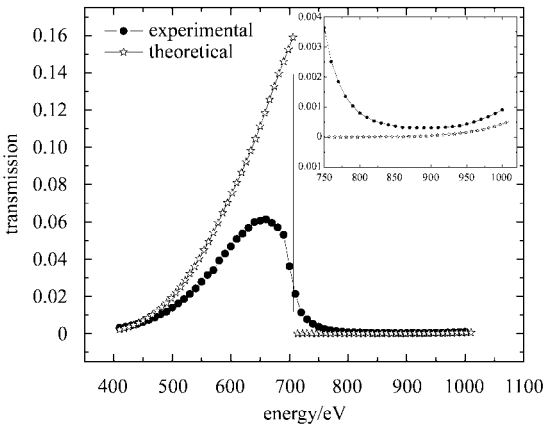

Fig.2. The measured transmission versus photon energy for a thin Fe film.

In this energy region, the uncertainty of the energy calibration of the beamline 3W1B is estimated to be 3%. It means that the energy reproducibility at any time is about 3% or better at higher energy where energy resolution decreases[3]. Bacause a low resolution broaden the experimental features at the absorption edge, data are usually not so sharp as theoretical curves[19] at the edge, as shown in Fig.2. Moreover, in the edge region, e. g., the energy range 707—801eV, the photo-absorption curves are in poor agreement with theory because the model used does not take into account the local structure contributions. The difference between experimental transmission data and theory decreases gradually as the photon energy increases (see inset of Fig. 2) reducing to a Fig. 2 at photon energy of 1000eV.

3 Determination of the photoabsorption cross section

The energy dependent photo-absorption cross section σ (m^2) of a thin film can be obtained by the Beer law:

$$I = I_0 e^{-n\sigma z}, \tag{1}$$

and as mentioned above, the expression of the transmission of a thin film is:

$$T = \frac{I}{I_0}, \tag{2}$$

then it can be shown from Eqs. (1) and (2) that σ:

$$\sigma = \frac{1}{nz}\ln\left(\frac{1}{T}\right), \tag{3}$$

where n is the atomic density, z is the thickness of the thin film and T is the energy dependent transmission of a film and as shown in Figs. 1 and 2.

4 Results

The photoabsorption cross section σ for an Al film derived from Eq. (3), in the soft X-ray region 58—80eV, is plotted in Fig.3 as solid circle dots with error bar. A better determination of σ in this energy range has been pointed out by Smith[20] in its review of the optical constants of Al. Looking at Fig.3 it is evident that data are almost constant and about 2.6 times larger than theoretical values in the energy range 61—72eV. Previous experimental data by Smith et al[20]. and Shile et al[21]. are also a factor of two larger than theory and both smaller than our data for energy below the edge. In addition only two experimental points are given by reflectance experiments of Windt[22] in this energy range. Above the edge, all measured data are lower than theory.

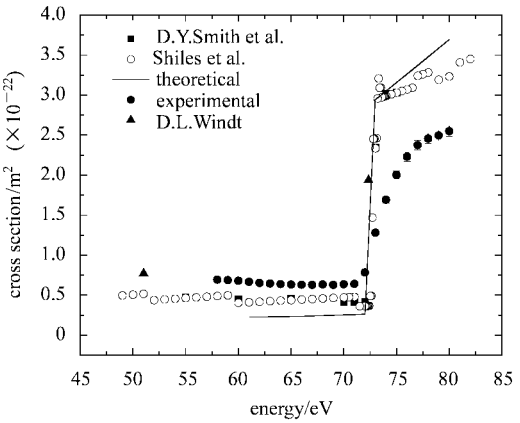

Fig.3. The photoabsorption cross section versus photon energy for a Al film.

In Fig. 4 we shown the experimental photo-absorption cross section (solid circles dots with error bars) of the Fe film

determined using the Eq. (3) in the energy region of 410—1010eV using steps of 10eV according to the available energy resolution. Previous experimental data are due to Palik[23].

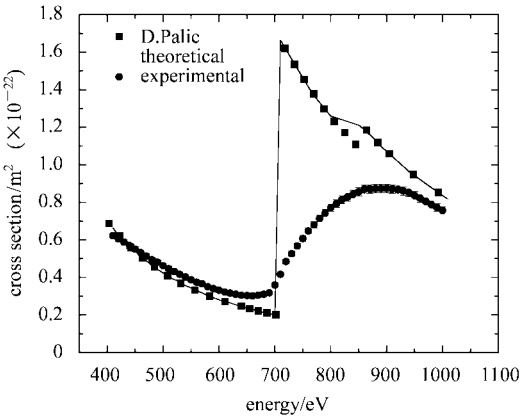

Fig.4. The photoabsorption cross section versus photon energy for a Fe film.

Below the Fe edge our data are in good agreement with Palik results while a poor agreement is observed above the edge. For these energies a large discrepancy also occurs between measured data and theory. Similarly to the Al case, the measured cross section is higher below Fe $L_{2,3}$ absorption edge and lower than theoretical data above the edge, probably because of the contributions of the stray light and of the high-order harmonics present in the incident beam. However, below the $L_{2,3}$ edge experimental data exhibit a better agreement respect to the Al film data. In addition, the experimental curve at near edge region in Fig. 4 is much broader than theoretical data, probably due to the lower energy resolution and the roughness of film surface.

Let us now discuss the uncertainty of the experimental cross section. The relative thickness uncertainty is about 1% for the Al film and 0.7% for the Fe film and the signal from the electrometer is accurate within 1%[24]. Moreover, the density of a material in the form of thin film may be different from the bulk density, so that the atomic density n of Al and Fe film calculated using the bulk density is accurate within 2%. Taking in considerations the experimental relative uncertainty, cross section data can be estimated with an error of 2.55% (rms) in the entire energy range. However, the uncertainty due to the presence in the incident monochromatic beam of radiation of different energies is larger than this error. At present, the beam purity at 3W1B is not determined[17] and a systematic error may affect our data.

5 Conclusions

Energy dependent photo-absorption cross sections for Al, in the energy region 58—80eV, and for Fe, in the energy region 410—1010eV have been measured at the beamline 3W1B at BSRF, and data have been compared with previous published experiments. The cross section for Fe is in good agreement with theory for energy above the $L_{2,3}$ edge, but only in qualitative agreement in the near edge region. Data for Al are about 2.6 times higher than theory in the range 58—72eV and 0.6 times higher in the range 75—80eV. This research confirm that cross section determinations in the transmission mode are possible also for thin films, although large discrepancies between experiments and theory have been observed for Al and Fe films. The independent atom approximation, on which theoretical values are calculated, is certainly not valid for photon energy near the absorption edge, and comparison between photo-absorption cross sections in this region is not clear. The experimental error of our data is estimated better than 3%, however, to improve the experimental determination it will be necessary to improve the characterization of the beamline 3W1B. A detailed evaluation of the content of high-order radiation and of the stray light in the monochromatic beam may improve experimental accuracy and enhance the energy resolution. In addition, it is necessary to improve the method to measure the film thickness.

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基于同步辐射软 X 射线能区的 Al 和 Fe 薄膜光吸收截面的初步研究*

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摘要　利用北京同步辐射装置(BSRF)3W1B 光束线产生的单色软 X 射线分别测定了 Al 和 Fe 薄膜在 $L_{2,3}$ 吸收边附近的光吸收截面.测量值与理论值比较在远离吸收边时偏差较小,并逐渐趋于一致,在吸收边附近有较大偏差,这主要归因于理论计算所采用的独立电子近似方法在吸收边附近是无效的.测量结果与若干文献中的实验结果也进行了对比.虽然由于光束线分辨率和高次谐波的影响,实验结果和理论结果在实验能区有一定的偏差,但测量结果说明用薄膜透过率测定金属薄膜光吸收截面的方法是可行的.

关键词　光吸收截面　Al　Fe 薄膜　软 X 射线　同步辐射

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