

Determination of the refractive index at soft X-ray resonances

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Refractive index of vanadium determined by resonant diffraction of soft x-rays

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Abstract

The dispersive part of the refractive index of vanadium is determined by measuring the angular displacement of the first order diffraction peak of a V/Fe superlattice. The measurements were made using elliptically polarized synchrotron radiation which was scanned through the V $L_{2,3}$ absorption edges for different incident scattering angles. This x-ray scattering technique provides access to the direct determination of the dispersive part of the refractive index across an absorption resonance and to the change of values from below unity to above unity.

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I. INTRODUCTION

It is well known that for condensed matter, the real part of the refractive index for x-rays is slightly less than unity, that is to say x-rays are refracted in a direction slightly away from the surface normal. [1] However, in the vicinity of absorption edges (regions of anomalous dispersion) the refractive index is strongly modified and the real part may even become greater than unity. Detailed knowledge of the dispersive and absorptive contributions or, alternatively, anomalous scattering factors, are required for the design of nano-engineered synthetic thin film materials. [2] This is especially the case in the soft x-ray region where wavelengths may be comparable to layer thicknesses or multilayer periods. Suitable data may be deduced from the absorptive part of the refractive index by applying the Kramers–Kronig transformation and have been tabulated for most elements (see Ref. [3]). Ideally, photoabsorption coefficient measurements use thin foils in the transmission mode. However, in the soft x-ray region it is often difficult to make sufficiently thin, free-standing films for transmission measurements and photoelectron yield techniques are used with bulk materials instead. The small depths probed because of the small mean-free path of the photoelectrons (at best a few tens of Å) are a handicap in general because of the risk of contamination and make the technique unsuitable for describing thick multilayers and buried interfaces. [4] Another suitable method is to measure specular reflection because this all-photon technique is less surface sensitive, though glancing angles are generally used to work in the region of total reflection. [5]

A more powerful technique developed recently is resonant Bragg diffraction performed in the neighborhood of a photoabsorption edge using a periodic structure such as a superlattice. It is then possible to obtain the dispersive part of the atomic scattering factor, or, alternatively, the refractive index, without using the Kramers–Kronig transformation. [6,7] In Ref. [8], a V/Fe superlattice was used to measure the dispersive part of the refractive index of Fe in the region of the Fe $2p$ -edge and its dependence on magnetization.

The bulk sensitivity and element selectivity of the x-ray Bragg diffraction technique make

it possible to measure a capped sample fabricated *ex situ*. Here we apply the technique to the determination of the refractive index of elemental vanadium in the vicinity of the V $2p$ -edge and compare the results with tabulated values obtained by the total electron yield technique. We also examine the need for applying absorption and polarization corrections.

II. EXPERIMENTAL DETAILS

The x-ray Bragg diffraction measurements were performed using the soft x-ray metrology bending-magnet beamline 6.3.2 at the Advanced Light Source (ALS). [9,10]

The sample was epitaxially grown by dual-target magnetron sputtering deposition of metallic V and Fe layers onto a polished MgO(001) fcc single crystal substrate at 300°C. [11] The alternating depositions of the V and Fe layers were repeated to form a total of 40 periods and capped with a Pd film to prevent oxidation. The structural quality of the sample was characterized by conventional $\theta - 2\theta$ x-ray diffraction (XRD) measurements with Cu $K\alpha$ radiation for low angles (1–14° in 2θ) and high angles (50–80° in 2θ) around the Fe/V (002) Bragg peak. The thickness parameters were obtained by a refinement procedure to reproduce the Bragg peaks of the XRD data using the simulation program SUPREX. [12] The periodicity $\Lambda = t_1 + t_2$ was determined to be ≈ 19.7 Å and the individual thicknesses of Fe ≈ 7.4 Å (6 ML) and V ≈ 12.3 Å (7 ML). The Pd capping layer was estimated to be ≈ 40 Å thick.

The incident photons were $\approx 60\%$ circularly polarized. The sample was mounted in the reflectometer endstation with the axis of rotation parallel to the orbit plane. The monochromator was set to a resolving power of about 2000 for a flux of $\approx 10^{10}$ photons/second on the sample at the V L_3 -edge. The magnetism induced by the Fe layers was found to be very small. Thus the curves shown here are the average of each magnetization directions relative to the polarization of the incoming photons.

III. RESULTS AND DISCUSSION

Figure 1 shows the resonant $L_{2,3}$ first-order diffraction peak of vanadium measured as a function of incident angle and as a function of photon energy. Intensities are normalized to the same incident photon flux. Variations in the intensity and width of the Bragg peak are observed when approaching the V L_3 and L_2 edges situated at 514.0 eV and 520.8 eV respectively. Refraction changes the angle of propagation of the radiation entering the multilayer and therefore changes the angular position of the Bragg peak. The value is $\approx 37.5^\circ$ in the vicinity of the L_3 edge. The width of the peak increases significantly compared to the off-resonance value as a consequence of the increased absorption which reduces the number of participating superlattice planes scattering in phase.

Figure 2 shows the angular distribution of the Bragg peak as a function of photon energy. The characteristics of the Bragg peak in terms of position, width, and intensity were extracted by standard interpolation and fitting procedures. The dashed line represents the wavelength dependence of Bragg's law $\theta_B = \arcsin(m\lambda/2\Lambda)$, where m is the order of diffraction, λ is the wavelength, and Λ is the periodicity of the multilayer. The large angular displacement around the $L_{2,3}$ resonances is directly related to the energy dependence of the refractive index in that region. The complex refractive index is $n = 1 - \bar{\delta} - i\bar{\beta}$ where $\bar{\delta}$ and $i\bar{\beta}$ are the average dispersive and absorptive correction terms respectively.

Assuming that absorption is negligible ($\beta=0$), we can write, from Bragg's law, that

$$m\lambda = 2\Lambda \sin \Theta \sqrt{\left(1 - \frac{2\bar{\delta} - \bar{\delta}^2}{\sin^2 \Theta}\right)} \quad (1)$$

with $\bar{\delta} = \gamma\delta_1 + (1 - \gamma)\delta_2$, where subscripts refer to different materials and γ is the division parameter defined by the thickness ratio of the layers with respect to the periodicity: $\gamma = t_1/(t_1 + t_2)$. Here $\gamma = 0.6$. If Λ is known, measurements of the angular positions of the Bragg peaks as a function of energy may be used to determine $\bar{\delta}$. If the individual thicknesses t_1 and t_2 , and thus the composition, is known the unit decrements of the dispersive parts of the refractive indices δ_1 and δ_2 of the component materials can also be determined. Since

$\bar{\delta} \ll 1$ in the soft x-ray region, the quadratic term in Eq. 1 can be neglected and we can write

$$\delta_1 = \left[\sin \Theta \left(\sin \Theta - \frac{p\lambda}{2\Lambda} \right) - \delta_2 \right] / \gamma + \delta_2. \quad (2)$$

Clearly the effect of absorption in the soft x-ray region must be included and the full correction can be written by replacing γ by D (Ref. [14]) where,

$$D = \gamma - \frac{(\beta_1 - \beta_2)\sin^2(m\pi\gamma)\bar{P}^2(\theta)}{m^2\pi^2[\gamma\beta_1 + (1 - \gamma)\beta_2]}. \quad (3)$$

For the geometrical set-up used in our experiment the polarization dependence $\bar{P}^2(\theta)$ would have been independent of θ for in-orbit-plane linear polarization (*s*-polarization). The *p*-component of $\bar{P}(\theta)$ has a $\cos(2\theta)$ dependence. At $\theta = 39.5^\circ$ the correction needed to account for the angular dependence of *p*-polarization component is completely negligible. From Eq. 3 we see that correction for absorption as a function of energy is not negligible. β_2 is essentially independent of energy as it corresponds to the absorption below the Fe *2p*-threshold. β_1 can be estimated from x-ray absorption measurements. Here we have used the values reported by Harp *et al.* [16] and normalized them to the tabulated values over the energy region 490–530 eV.

Figure 3 shows the dispersive part of the refractive index $(1 - \delta)$. Tabulated values by Henke *et al.* [3] are shown for comparison as well as the Kramers-Kronig transformation of the absorption data by Harp *et al.*

Below the $L_{2,3}$ absorption edges, the refractive index is, as expected, less than unity and slowly monotonically increases as a function of energy in accordance with normal dispersion. At about 501 eV, the refractive index becomes larger than unity and increases rapidly up to a peak value of 1.0079 at the L_3 edge at 514.0 eV and 1.0045 at 520.4 eV at the L_2 edge. Following each absorption edge, the refractive index drops back sharply to values less than unity as a consequence of the anomalous dispersion effect.

The tabulated refractive index values reported in the literature [3] are in reasonably good agreement with our measurements considering that they do not resolve the spin-orbit

splitting. The more recent x-ray absorption data reported by Harp *et al.*, [16] however, are in excellent agreement though it is likely that the better agreement with our uncorrected curve rather than with the corrected curve is fortuitous. The values we have measured are certainly representative of bulk values despite the relatively small thickness of the layers so the comparison with the tabulated values is fully justified. The difference between the measured and tabulated values is shown up as smaller positive values of n and the absence of dips in n just above each of the two $2p$ edges. In the energy region above ≈ 520 eV the absorption of V is large, the average refractive index $1 - \bar{\delta}$ is close to that of Fe which implies that n does not strongly depend on γ above the $VL_{2,3}$ thresholds.

The effect of the absorption correction on the refractive index of vanadium depends appreciably on the photon energy. For energies below the L_3 threshold, the factor D decreases from 0.67 to 0.65 which is larger than the nominal γ -value of 0.6 and implies a correction towards lower refractive index. In the energy region ≈ 514 – 530 eV the factor D drops to values of the order of 0.47–0.50 implying a correction towards higher refractive index as shown in Fig. 3.

The absorption correction is significant in the energy region of the $L_{2,3}$ edges. However, it is small for photon energies above the $2p$ absorption edges due to the fact that n does not strongly depend on γ , and therefore D , in this energy region. The effect of polarization on the magnitude of the absorption correction is negligible since the measurements were performed close to the Brewster angle.

For x-ray optics such as mirrors, it is often desirable to maximize the critical angles and therefore absorption thresholds are generally to be avoided. However, tuning grazing incident photons to energies close to an absorption threshold where the real part of the refractive index is larger than unity will induce total internal reflection and standing waves along the interface of a multilayer. Like its counterpart in the visible region, total internal reflection of x-rays over a limited bandwidth could be one of the uses of such multilayer structures.

IV. CONCLUSIONS

The dispersive part of the refractive index, $1 - \delta$, of vanadium was determined by measuring the angular dependence of the Bragg peak at the V $L_{2,3}$ edge energy region using a V/Fe superlattice. We demonstrate that previously tabulated values tend to underestimate the amplitude of the change in the real part of the refractive index. This illustrates the importance of performing direct measurements to determine the optical properties of multilayer devices. In the soft x-ray energy region where absorption effects are significant, corrections to take into account absorption and polarization effects have been assessed.

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FIGURES

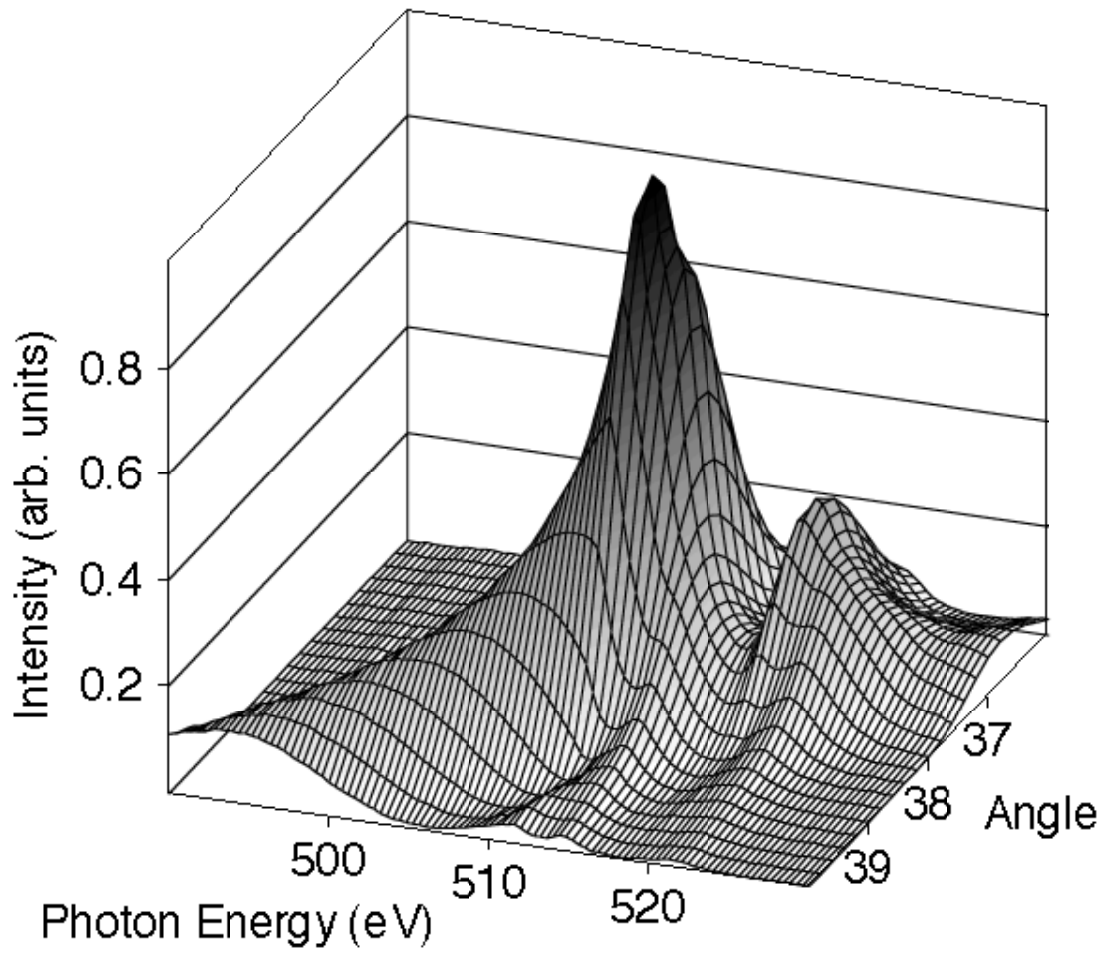


FIG. 1. The first-order Bragg peak measured by scanning photon energy through the V $L_{2,3}$ absorption resonances as a function of angle.

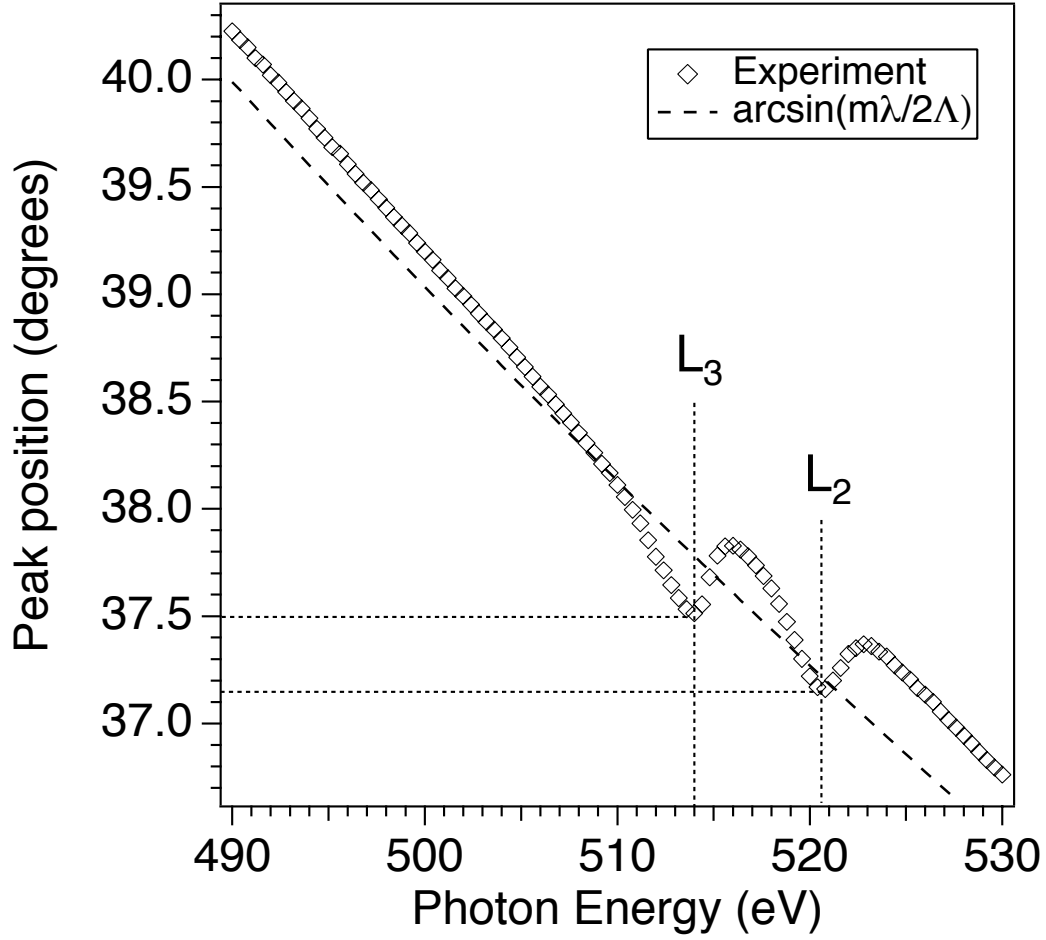


FIG. 2. Angular position of the Bragg peak maximum as a function of photon energy at the $2p$ edges of vanadium. The sloping dashed line is obtained from Bragg's law. The dotted lines indicate the angular positions of the intensity maxima (37.51° at 514.0 eV and 37.16° at 520.8 eV at the L_3 and L_2 thresholds, respectively).

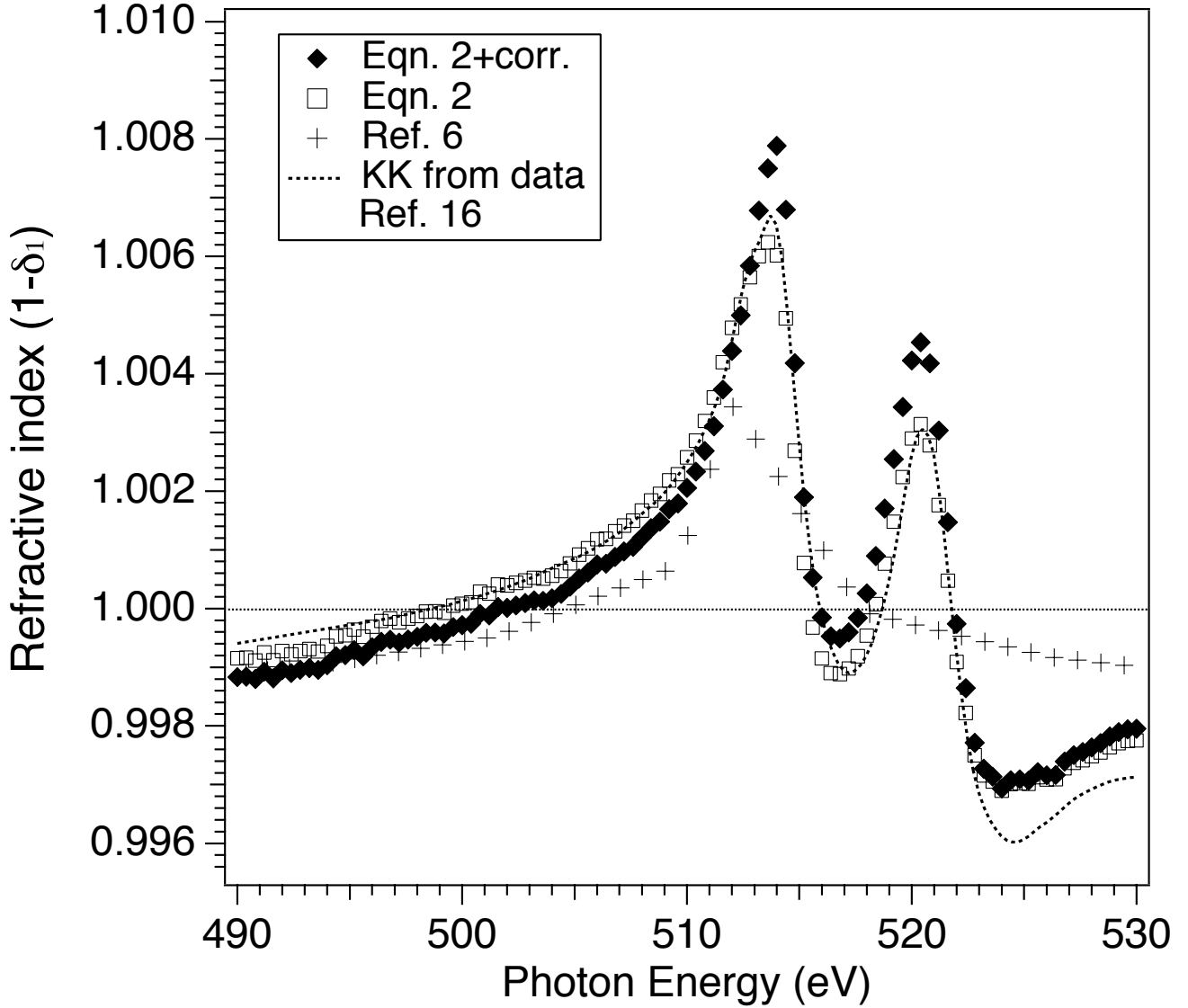


FIG. 3. Energy dependence of the real part of the refractive index, $1 - \delta$, of vanadium at the $V L_{2,3}$ edge. Also shown is the energy dependence in the absence of absorption correction, the tabulated values due to Henke *et al.* [3] and the Kramers-Kronig transformation of the absorption data by Harp *et al.* [16].