Optical Constants of Ferromagnetic Iron via $2p$ Resonant Magnetic Scattering

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We have determined the optical constants at the $2p$ edges of iron by measuring the Bragg scattering from a Fe/V superlattice using elliptically polarized photons whose energies were tuned across the Fe $2p$ resonances (700–730 eV). The analysis of the Bragg peak displacement as a function of photon energy and sample magnetization, together with standard absorption measurements, allowed to determine the complete dielectric tensor for Fe, including its off-diagonal terms. [S0031-9007(98)06882-3]

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For decades, extended or near-edge x-ray absorption spectra have been used to determine local structure and chemical environment element specifically [1]. Over the last ten years, the use of polarized x rays has widened this field to include the study of anisotropic systems, and, in particular, magnetic materials [2] with, at the forefront, interest in tailor-made magnetic multilayers. Modeling these requires simultaneously taking into account the spin polarization of the conduction electrons, changes in the dielectric constant at interfaces, and interlayer magnetic coupling. Confrontation with experiment, at this stage of the development of first principles calculations, is imperative. The energy dependence of the optical constants at a resonant excitation represents a privileged standpoint for such a confrontation [3], yet in the x-ray region only the imaginary part β (i.e., absorption) of the complex refractive index $n = 1 - \delta - i\beta$ is used. Model calculations, on the other hand, may deal with the real and imaginary parts on the same footing. The problem is simply an almost complete lack of experimental data even for standard materials, let alone for multilayer structures. While it is true that the real part of the index provides similar information to the imaginary part [the former may be calculated from the latter via a Kramers-Kronig (KK) transformation], it contributes a further set of *independent* parameters for refining theoretical models [4]. It has even been shown that interference between real and imaginary parts (as in resonant reflectivity) can facilitate the investigation of spectroscopic features, such as satellites, that are weak but of major importance for understanding some ground state properties [5].

Most of the few direct experimental determinations of δ at resonance (i.e., without using KK relations) have been performed on crystals by measuring the Bragg peak displacement when the photon energy is scanned through an absorption edge [6]. The Bragg law imposes the relation between the photon wavelength λ and the crystal spacing $2d$ (a few Å). This explains that x-ray resonant magnetic scattering (XRMS) experiments have been performed mainly at high energies (hard x rays) [7]. However, x-ray magneto-optics effects, related to the off-diagonal terms of the dielectric tensor, are stronger when the core excitations produce transitions to a final state that directly involves the *magnetic* orbitals [$3d$ for the first row transition metals (TM) or $4f$ for rare earths (RE)]. The largest cross sections are observed for dipolar transitions, which implies that the most interesting resonances for studying magneto-optics effects [$(2,3)p \rightarrow 3d$ for TM and $(3,4)d \rightarrow 4f$ for RE] are all located in the so-called soft x-ray region (below 2 keV). This means that metallic multilayers with artificial periodic structures with $2d$ spacings in the range 10 to 200 Å are ideally suited to performing XRMS with soft x rays [8,9].

In this Letter we show that the real part δ of the index of Fe across its $2p$ resonances (dominated by $2p \rightarrow 3d$ transitions) may be determined by analyzing the Bragg diffraction from a metallic superlattice constituted of alternate Fe and V layers deposited on MgO. For this experiment we chose a system where Fe is known to have closely the same magnetic properties as in bulk form [10]. A future step will be to correlate magneto-optics constants and magnetic properties in multilayer devices presenting specific magnetic anisotropies as a function of environment (e.g., metastable phases stabilized by epitaxial growth [11]).

Our results are discussed in relation to the only other direct determination of δ obtained by measuring the Faraday

rotation of x rays transmitted by a thin film of ferromagnetic iron [12]. We also confront them to a KK transformation of the dichroic absorption recorded simultaneously on our sample.

The sample is an epitaxially grown Fe/V superlattice prepared by sputter deposition of the metallic layers on an MgO (001) crystal [13]. Alternate depositions of Fe and V layers (nominally five atomic layers each) were repeated to form a 40 period superlattice. Hard and soft x-ray reflectivity measurements gave a $2d$ spacing of 30.6 Å and a value of 0.50 ± 0.01 for the division parameter γ, defined by the thickness ratio $t_{\mathrm{Fe}}/(t_{\mathrm{Fe}} + t_{\mathrm{V}})$. The sample was capped by 18 Å of Pd in order to prevent oxidation. The superlattice was characterized by high and low angle x-ray diffraction, showing good crystal quality and a mean interface roughness lower than 2 Å [13]. The easy axes of magnetization were determined to be oriented along the [110] axes of the substrate, i.e., [100] in-plane directions of the superlattice.

X-ray scattering and absorption measurements were performed on the soft x-ray metrology beam line 6.3.2 at ALS (Berkeley) [14], using the radiation emitted by a bending magnet between 110 and 200 μrad above the orbit plane, which gives elliptically polarized photons with a circular polarization rate of 40% over the 650–780 eV energy range. The polarization state of the light is preserved by the grazing incidence optics of the beam line. The dispersive element is a plane grating with varied line spacings, giving (for the adopted exit slit aperture of 50 μm) a resolving power of 2000 at 700 eV and a flux of about 10^{10} photons per second on the sample. The experimental setup consists of a $\theta/2\theta$ reflectometer which allows xyz adjustment of the sample position. The sample was magnetized along an easy axis, parallel to its surface and in the scattering plane, by means of a permanent magnet situated behind the sample holder and mounted on a stepper motor used to reverse the field direction. The field at the sample surface was 800 G, i.e., well above the coercive field. $\theta/2\theta$ scans were performed over the 33°–38° range in steps of $\Delta\theta = 0.05°$ and for photon energies between 700 and 730 eV, with a step of 0.2 eV at the L_3 edge and 0.6 eV at the L_2 edge. The incoming flux was monitored and used to normalize the spectra. Collecting the resonant magnetic scattering data reported here took approximately 8 hours. Absorption spectra were recorded by measuring the drain current from the sample. An angle of incidence of 30° was used in order to minimize saturation effects [15].

Resonant scattering spectra are reported in Figs. 1(a) and 1(b) as a function of θ and photon energy for the two magnetization to helicity orientations. These are raw data, simply normalized to the incoming photon flux. Figure 1(c) shows the difference between curves (a) and (b).

A preliminary comment should be made concerning the photon energy dependence of the Bragg scattering intensity: it shows a strong variation when approaching the L_3 and, to a lesser extent, the L_2 edges of Fe. This variation depends on the magnetization of the sample, as is clearly seen in Fig. 1(c). To better appreciate this energy dependence, Fig. 2(a) shows the integrated peak intensity versus photon energy for the two magnetization directions: a change of 2 orders of magnitude is observed over a range of less than 10 eV across the L_3 edge. This effect is well known and has been discussed in the past [6,8,16]. On the one hand, it is related to the large variation in δ at resonance. This changes, as we will discuss below, the effective electronic density of Fe for the scattering process as a function of the photon energy and consequently increases the contrast with the nonresonant V. On the other hand, the increasing absorption at the Fe edges shortens the penetration of the photons in the sample and reduces the number of planes capable of scattering in phase, resulting

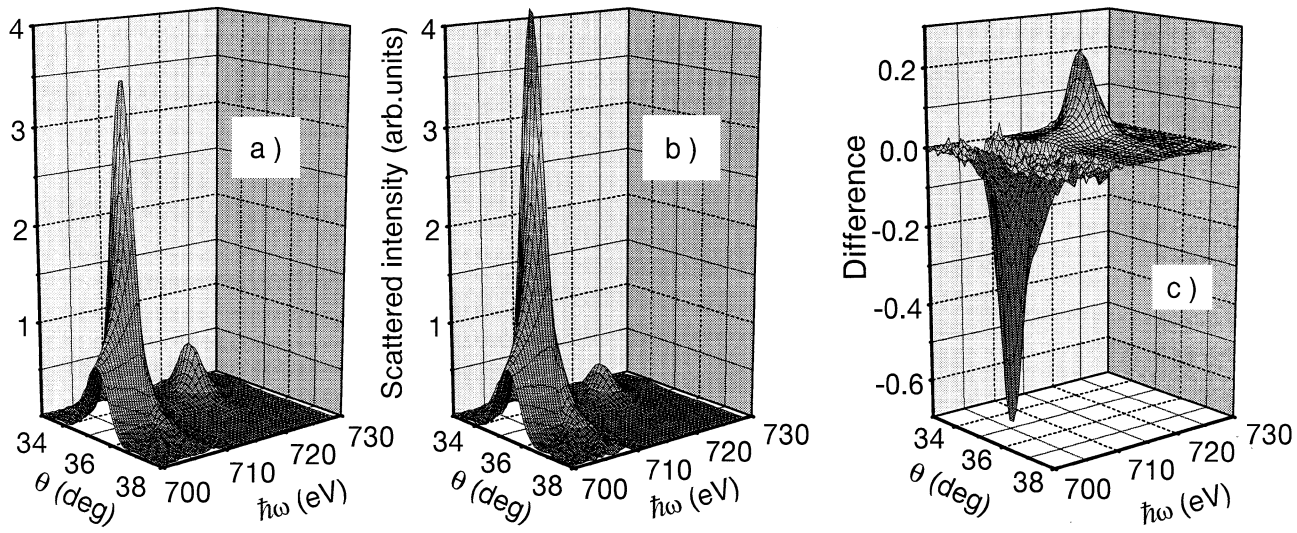

FIG. 1. $\theta/2\theta$ scans at various photon energies ($2p$ edges of Fe) for (a) parallel and (b) antiparallel magnetization/helicity orientations. Panel (c) reports the corresponding difference curves.

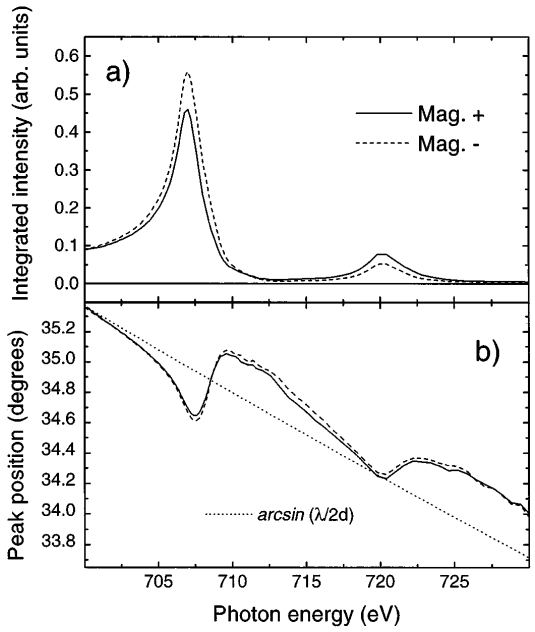

FIG. 2. Bragg peak of the superlattice as a function of photon energy. (a) Integrated intensity. (b) Angular position. In both panels, continuous and dashed lines refer to opposite magnetizations of the sample along a [100] direction.

in broader and weaker peaks. A fit of the data with a Voigt function of fixed Gaussian contribution indicates a Lorentzian broadening that varies by a factor of 10 across the L_3 edge of Fe. Figure 1 also shows the displacement of the maximum of the Bragg peak position θ_M as a function of photon energy. This is partly related to the change in wavelength, but, as shown in Fig. 2(b), large deviations are observed with respect to a simple arcsine behavior. These deviations are related to the energy dependence of δ which is something we will bring out in what follows. It is apparent, moreover, that there is a difference in the curves for opposite magnetizations [Fig. 2(b)], giving us access to the magnetic part of δ for Fe.

A simple way of obtaining δ from the data in Fig. 2(b) is to consider the average decrement given by

$$\overline{\delta} = \gamma\delta + (1 - \gamma)\delta_{\mathrm{V}} = \left(\sin\theta - \frac{\lambda}{2d}\right)\sin\theta\,, \qquad (1)$$

where the subscript V relates to the optical constants of vanadium. Equation (1) accounts for refraction effects, but is only valid for a real index. A further correction term should be introduced to account for the effect of absorption on the Bragg peak position. Following the model of Rosenbluth and Lee [17] one has

$$\delta = \left[\left(\sin\theta - \frac{\lambda}{2d}\right)\sin\theta - \delta_{\mathrm{V}}\right] \Big/ D + \delta_{\mathrm{V}}\,, \quad (2)$$

where

$$D = \gamma - \frac{(\beta - \beta_{\mathrm{V}})\sin^2(\pi\gamma)}{\pi^2[\gamma\beta + (1 - \gamma)\beta_{\mathrm{V}}]}\,. \qquad (3)$$

To determine δ from Eqs. (2) and (3) we have scaled the experimental absorption spectra to the calculated values over the 650–680 eV and 750–780 eV energy ranges, i.e., below and above the $2p$ resonances. Absorption correction is found to have little, but not negligible, effect on δ for Fe. Finally we have corrected the δ and β curves for the incomplete polarization of the light, and the β curves also for the incomplete alignment between the photon propagation axis and the magnetization. The results are shown in Fig. 3(a). The four curves are all we need to construct the dielectric tensor ϵ for Fe over the 700–730 eV energy range. ϵ has elements

$$\epsilon_{yz} = -\epsilon_{zy} = -i(\epsilon^{+} - \epsilon^{-})/2\,,$$

$$\epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = (\epsilon^{+} + \epsilon^{-})/2\,,$$

$$\epsilon_{xz} = \epsilon_{zx} = \epsilon_{xy} = \epsilon_{yx} = 0\,,$$

where x is the magnetization axis and $\epsilon^{\pm} = (1 - \delta^{\pm} - i\beta^{\pm})^2$. The equality of the diagonal elements relies on the hypothesis that cubic Fe does not show any linear dichroism. Otherwise additional measurements with linearly polarized light along the main axes of symmetry would be required.

In summary, we have obtained experimentally the optical constants of Fe over the 700–730 eV energy range covering the $2p$ resonances. Resonant magnetic scattering from a superlattice makes it possible to obtain the real part of the refractive index without using KK transformations. In a previous work, Kortright *et al.* [12] used another method to the same end; i.e., they measured the Faraday rotation of the polarization upon transmission through a thin magnetized Fe layer. Both techniques

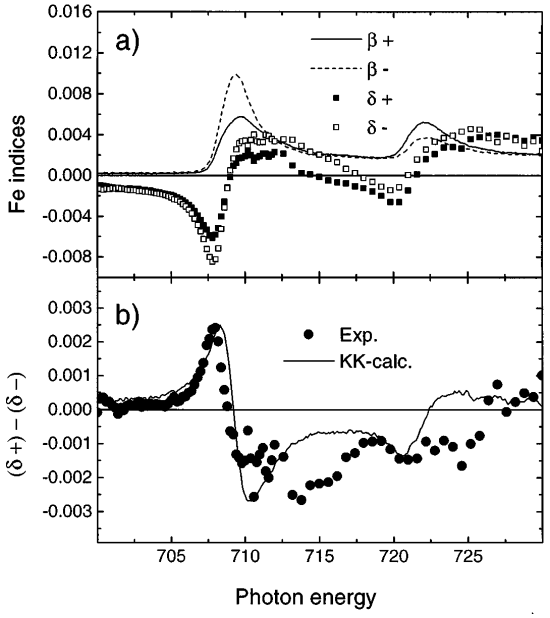

FIG. 3. (a) Imaginary part (β) and decrement of the real part (δ) of the complex refractive index of Fe. Plus and minus signs refer to opposite magnetization orientation. (b) Comparison between $(\delta^{+} - \delta^{-})$ and the KK transformation of $(\beta^{+} - \beta^{-})$.

present some experimental difficulties and require a certain degree of data reduction, but we want to stress that, even at this stage where they are not yet optimized, both give access to information difficult to attain with the same reliability by other methods.

As far as comparisons with the previous results are possible, we can say only that we observe a much larger magnetic component of δ than reported in Ref. [12]. Figure 3(b) shows the magnetic part of δ, which, if expressed in electrons as in Ref. [12], gives values as large as 20 electrons around 707–708 eV, i.e., at the steep rise of the L_3 edge. Such a high value is consistent with the KK transformation of the dichroism in the imaginary part β, also reported in Fig. 3(b). Despite a general quantitative agreement, it might be objected that the scattering data are noisier than those obtained by KK transformation, but we stress again that the latter is influenced by hidden and less well controlled parameters such as arbitrary scaling to tabulated values and choice of approximate integration methods. Whenever the signal to noise ratio is such that a clear difference is observed, we should give more credit to the XRMS determination. This is the case in the 705–710 eV region [see Fig. 3(b)].

To conclude, this application of XMRS to the region of the Fe $2p$ edges is the first of the kind. It especially demonstrates feasibility and points the way to future developments where the advantages of a photon-in/photon-out experiment and a large probing depth are at a prime. We consider that the experimental determination of the dielectric tensor including its off-diagonal terms is an important step towards a better refinement of theoretical and computational models that describe the electronic and magnetic ground state properties of materials. To this end, it is essential to develop several independent experimental methods in addition to the standard KK approach. Polarization rotation (upon transmission or reflection), interferometry, and resonant magnetic scattering constitute together a sound basis of complementary techniques for future work.

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