Novel Interferometer in the Soft X-Ray Region


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A novel interferometer has been developed with which the complex index of refraction of thin self-supporting foils can be measured in the vacuum ultraviolet and soft x-ray region. It consists of two collinear undulators and a grating spectrometer. Taking advantage of the low emittance 855 MeV electron beam from the Mainz Microtron, distinct intensity oscillations have been observed as a function of the distance between the undulators. A foil placed between the undulators causes a phase shift and an attenuation of the oscillation amplitude. The complex index of refraction has been measured at the K-absorption edge of carbon. [S0031-9007(98)06368-6]

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Resonant anomalous x-ray scattering plays an increasingly important role in many disciplines of physics, biology, and material sciences. Using the brilliant and tunable x-ray beams from modern synchrotron radiation sources, it is now possible to fully exploit the information in the strong energy and polarization dependence of the atomic scattering amplitude \( f(\omega, q) = f_0(q) + f'(\omega) + if''(\omega) \) near absorption edges [1]. This is the ratio of the scattering amplitude of an atom to that of a free (Thomson) electron. It can be separated in an energy independent “normal” coherent scattering amplitude \( f_0(q) \) and an “anomalous” or resonant scattering amplitude \( f'(\omega) + if''(\omega) \). It is common practice to assume that the resonant scattering amplitude does not depend on the momentum transfer \( q \) [2]. This assumption permits its measurement in forward scattering geometry. The imaginary scattering factor \( f'' \) can be directly determined from the total photon cross section \( \sigma(\omega) \) by employing the optical theorem: \( f''(\omega) = \omega \sigma(\omega)/4\pi r_0 c \). The total cross section is well approximated by the absorption cross section. The real part \( f' \) can be calculated from \( f'' \) by means of dispersion relations. The Cauchy principle value integral connects real and imaginary parts: They are a Kramers-Kronig or Hilbert transform pair. However, this method has an inherent drawback since it is an indirect method which requires precise absorption data for all frequencies from zero to infinity. Kramers-Kronig data are suited, therefore, for a relative comparison only [3]. If precise absolute values are needed, a direct measurement of \( f'(\omega) \) is required. Direct measurements are based on x-ray interferometry [4], refraction through a prism [5,6], diffraction from perfect crystals and pendellösung fringes [7,8], determination of the angle of total reflection [3,9], and Fresnel bimirror interferometry [10]. In the energy range above 50 eV and below about a few keV, the only available methods are the last two. Although total reflectivity has been proven to be a powerful method, there are accuracy limiting factors which originate from surface oxides or carbonaceous deposits, surface roughnesses and inaccurate knowledge of the atomic density at the surface [3]. Therefore, independent and accurate methods for the measurement of \( f' \) as the Fresnel bimirror interferometry are highly desirable in this energy range. They would not only be of interest in, e.g., mirror fabrication for synchrotrons, astrophysical devices, or for high temperature plasma diagnostics but also for x-ray lithography.

In this Letter a novel interferometer is described which consists of two undulators and a grating spectrometer. The performance of the interferometer has been demonstrated at the Mainz Microtron (MAMI) with measurements at the K-absorption edge of carbon at 284 eV. The dispersion \( \delta(\omega) = (1/2)(\omega_p/\omega)^2[f_0(0) + f'(\omega)]/Z \) and the absorption term \( \beta(\omega) = (1/2)(\omega_p/\omega)^2f''(\omega)/Z \) of the complex index of refraction \( n(\omega) = 1 - \delta(\omega) - i\beta(\omega) \) have been determined simultaneously for a 65 \( \mu \)g/cm\(^2\) amorphous carbon and a 35 \( \mu \)g/cm\(^2\) polyimide foil. In these expressions \( \omega_p \) is the plasma frequency with \( \omega_p^2 = 4\pi r_0^2 n_a Z \), \( r_0 \) the classical electron radius, \( n_a \) the number of atoms per volume, and \( f_0(0) = Z \) neglecting relativistic corrections. A high accuracy has been reached, even in the region where \( \beta \geq \delta \) in which the optical constants can be determined from reflectance measurements only with large uncertainties [11].

The basic idea of the interferometer will be explained by means of the schematic experimental setup shown in Fig. 1. The fixed downstream undulator \( U_r \) is the reference undulator. The upstream undulator \( U_0 \) can be moved in the direction of the electron beam which coincides with the s axis. An electron with Lorentz factor \( \gamma \) traversing the two undulators experiences a sinusoidal magnetic field \( B_s = B_0 \cos(2\pi s/\lambda_U) \) perpendicular to the s direction and emits two electromagnetic wave trains. These are separated along the axis by the distance \( \Delta(\theta, d) \) which is a function of the distance \( d \) between the two undulators, and the observation angle \( \theta \) with respect to the electron beam direction. The grating spectrometer serves as a Fourier analyzer of the wave trains. The two resulting waves can approximately be described as plane waves \( A_r(\omega) \exp[-i(k s - \omega t)] \) and \( A_0(\omega) \exp[-i(k s - \omega t) + i\phi(\theta, d)] \) with the phase
difference $\phi(\theta, d) = \omega \Delta(\theta, d)/c$. Both waves interfere in the detector, and oscillations can be observed if the distance $d$ between the undulators is varied. The phase increases linearly with $d$ and the phasor $A_0(\omega) \exp[i \phi(\theta, d)]$ circles around the tip of the fixed reference phasor $A_0(\omega)$ resulting in an oscillation of the intensity $I(d) = |A_0(\omega) + A_0(\omega) \exp[i \phi(\theta, d)]|^2$. A foil of thickness $t_0$ placed between the undulators produces an additional phase shift $\phi_f = (\omega/c) \delta(\omega) t_0$ and attenuation $a(\omega) = \exp[-(\omega/c) \beta(\omega)t_0]$ with respect to the wave train from the undulator $U_r$. Consequently, both quantities can be extracted from the measured interference oscillations $I(d)$ with and without the foil between the undulators.

In a more rigorous description the intensity $I = |A|^2$ with a foil is given by

$$I = |A_r|^2 + |A_0|^2 e^{-(\omega/c) \beta(\omega)t_0} + 2|A_r||A_0| \exp[-(\omega/c) \beta(\omega)t_0] \cos(\omega/c[\Delta(\theta, d) + \delta(\omega)t_0]),$$

(1)

$$\Delta(\theta, d) = \frac{K_r^2}{4\gamma^2 L_U} + \frac{1}{2} \left( \frac{1}{\gamma^2} + \theta^2 \right) d.$$  

(2)

$K_r$ and $K_0$ are the undulator parameters with $K = (e/2m_e c) B_0 A_U = 0.934(B_0/T)(A_U/cm)$ the length of the undulator, $L_U$ the undulator period, and $n$ the number of periods. The amplitudes $A_r(\omega, \theta, K_r)$ and $A_0(\omega, \theta, K_0)$ depend via a function $x(\omega) = \pi n(\omega/\omega_0 - k)$ on the frequency $\omega$ with $k$ the harmonic number and $\omega_0 = (4\pi c^2 \gamma^2/L_u)/[1 + K_r^2/2 + (\gamma\theta)^2]$. They are given in a good approximation by the expression [12] $A(\omega, \theta, K) \propto [\gamma K/(1 + K_r^2/2 + (\gamma\theta)^2)] \exp(ix) \sin(x/\gamma)$ with the spectral function $\sin(x/\gamma)$.

The following orders of magnitude are expected in a realistic experiment. For an electron beam energy of 780 MeV, photons with an energy $\hbar \omega = 300$ eV ($k = 1$) are emitted from the undulators with the parameters given in the caption of Fig. 1. The wavelength of the interference oscillation $\lambda_d = (4\pi \gamma^2 c/\omega)/[1 + (\gamma\theta)^2]$, derived from Eqs. (1) and (2), amounts on axis ($\theta = 0$) to 19.26 mm. Using forward scattering amplitudes $f_0(0) + f^\prime(\omega) = 2.71$ and $f^\prime\prime(\omega) = 3.92$ [13] a 50 $\mu$g/cm$^2$ carbon foil, placed between the undulators, shifts the phase by $\phi_f = (\omega/c) \delta(\omega)t_0 = 0.79$ rad. This phase change corresponds to a distance change $\Delta d = 2.4$ mm. The amplitude is attenuated by a factor $a(\omega) = \exp[-(\omega/c) \beta(\omega)t_0] = 0.32$. Both values are large and can be measured with high precision.

Equation (1) is valid only for two identical undulators and under elsewhere ideal experimental conditions. In this case, the coherence $C$, defined as $C = \mu_{\max} - \mu_{\min}/(\mu_{\max} + \mu_{\min})$ with $\mu_{\max}$ and $\mu_{\min}$ the maximum and minimum intensities of the oscillation, reaches its maximum possible value $C = 1$. In a real experiment the angular beam spread or the finite accepted energy band of the monochromator, for example, causes phase smearings which may spoil the interference signal. Therefore, the most important sources attenuating the coherence will be evaluated in the following. The general expression for the coherence function is $C = 2|\langle A_r|A_0 \cos(\omega/c)\Delta \delta(\omega)t_0\rangle_{\mu_{\max}}|/|A_r|^2 + |A_0|^2$. The expectation values were calculated assuming that the various spreads can be well approximated by Gaussian distribution functions.

**Electron beam emittance.**—A detector with an infinitely small aperture positioned in the horizontal ($x, s$) plane at an angle $\theta_{s0}$ with respect to the $s$-axis, in a distance $D$ from the foil, views effective angular spreadsings $\Sigma^2_{x,y} = \sqrt{\sigma_{s,y}^2 + \sigma_{x,y}^2/(D^2)}$ in $x$ and $y$ direction if the waist of the electron beam is located in the foil. These spreads are caused by the horizontal and vertical emittances $\epsilon_{x,y} = \pi \sigma_{x,y} \sigma_{x,y}^\prime$ of the electron beam, with $\sigma_{x,y}^\prime$ and $\sigma_{x,y}$ the rms angular divergence and spot size, respectively. The expectation values of the coherence function were calculated using integral tables [14]. The result is

$$C(\omega, d, \Sigma^\prime_{x,y}, \Sigma_{x,y}, \theta_{s0}) = \exp[-(1/2)(S^\prime_{x,y} + (1 + S^2_{x,y}))/(\theta_{s0}/\Sigma_{x,y})^2] \sqrt{(1 + S^2_{x,y})/(1 + S^2_{x,y})}$$

(3)

with $S_{x,y} = (\omega/c) d \Sigma^2_{x,y}$. Equation (3) is a good approximation close to the maximum of the spectral function. The optimum coherence is reached for $\sigma_{x,y} = \sqrt{\epsilon_{x,y} D/\pi}$ resulting in $\Sigma^2_{x,y} = 2\epsilon_{x,y} D/(\pi D)$. With the requirement that (a) $C_D \geq \sqrt{1/2}$ for on-axis observation ($\theta_{s0} = 0$) and (b) $C_D \geq \exp[-(1/2) \sqrt{2}$ for off-axis observation ($\theta_{s0} > 0$), coherence conditions

$$\frac{d}{D} \frac{\epsilon_{x,y}}{\pi} \leq \frac{\lambda}{4\pi}$$

(4a)

$$\theta_{s0,y} d \frac{\epsilon_{x,y}}{\pi D} \leq \frac{\lambda}{2\pi}$$

(4b)

can be derived from Eq. (3), respectively. For the photon energy of 300 eV the coherence condition (4a) is fulfilled with the parameters as given in Fig. 1 and the emittances $\epsilon_x = 7\pi$ nm rad, $\epsilon_y = 1\pi$ nm rad available at the MAMI at 855 MeV beam energy. The coherence condition (4b) restricts the observation angles to
the rather small values $\theta_x = 0.95 \times 10^{-4}$ rad and $\theta_y = 1.78 \times 10^{-4}$ rad.

Small angle electron scattering.—The coherence function $C_{sc}(\Delta \sigma_{sc}, \sigma^I_{sc}, \theta_{sc})$ which originates from small angle scattering of the electrons in the foil is given by an expression of the same structure as Eq. (3) with $\sigma^I_{sc} = \sigma^I_y = \sigma^I_x$, and $d_f = 8.9$ cm the distance between the foil and the center of the reference undulator. The quantity $\sqrt{2} \sigma^I_{sc}$ is the rms value of the Gaussian distribution in space. $(\gamma \sigma^I_{sc})^2$ has been calculated with the formula (7) of Ref. [15] (note that a square on $\sigma$ is missing there) for a central fraction $F = 0.995$. For a 50 $\mu$g/cm$^2$ carbon foil a $(\gamma \sigma^I_{sc})^2 = 1.5 \times 10^{-4}$ is calculated and, at $\theta_{sc} = 0$, the coherence loss $\Delta C_{sc} = 3.8 \times 10^{-5}$ is negligibly small. However, associated with the scattering there is also a phase shift $\Delta \phi_{sc}(d_f) = -\Delta \sigma_{sc}(d_f)\sigma^I_{sc} = -\sqrt{2} \Delta \sigma_{sc}$ which amounts to 8.7 mrad. This value is in fact just one half of the accuracy achieved in the experiment described below.

Resolution of the spectrometer.—The coherence function $C_{s}(\sigma, \gamma_{sc})$, associated with the relative energy resolution $\sigma_{e}/\omega$ of the spectrometer, was calculated as

$$C_{s}(\Delta \sigma, \sigma_{e}) = \exp\left(-\frac{1}{2} \Phi_{s}(0, d) \sigma_{e}/\omega^2\right)$$

with the phase $\Phi_{s}(0, d) = (\omega/c)\Delta(0, d)$. Formula (5) is valid for a relative energy resolution $\sigma_{e}/\omega \ll 1/\pi k n$. At a still acceptable coherence loss $\Delta C_{s} = 0.1$ and the parameters given in Fig. 1, the FWHM resolving power $\omega/(2.355\sigma_{e})$ must be at least 134. To resolve fine structures in the anomalous dispersion it must be better.

Magnetic fields of the two undulators.—A difference $\Delta K$ between the undulator parameters $K_0$ and $K_1$ has only little influence on the coherence since $K$ is close to $\sqrt{2}$ for which the amplitude reaches a maximum. However, since the amplitudes $|A_0|$ and $|A_1|$ should not differ by more than 5% within the half–width of the spectral profile, a $\Delta K < 0.05$ is required for $k = 3$, the third harmonic. A static magnetic dipole field, e.g., at the entrance of the downstream undulator, results in a coherence loss $\Delta C_{B} = (1/2)\alpha^2(x) (\gamma \Delta \theta)^4$, with $\Delta \theta$ the deflection angle and $\alpha(x) = -[1 + (1/x - 1/\tan x)(x + \pi k n)]/(1 + K^2/2)$. For a still tolerable $\Delta C_{B} = 0.1$ the deflection angle must obey $\Delta \theta \leq 0.12/\gamma$, for $k = 3$, which means that any integral magnetic dipole component must be tuned away.

A coherence loss by the energy spread $\Delta \gamma$ of the electron beam can be disregarded since for MAMI a $\Delta \gamma/\gamma = 5.5 \times 10^{-5}$ (FWHM) has been measured at 855 MeV [16]. Transition radiation and bremsstrahlung contributions from the sample foil are estimated to contribute a negligibly small fraction of radiation in comparison with the undulator radiation. This discussion shows that the proposed interferometer can be realized with an electron beam as delivered by the Mainz Microtron. The experimental verification is described in the following.

Two 10-period hybrid undulators were constructed with identical periods $\lambda_U = 1.2$ cm employing NdFeB permanent magnets with a high remanence and a CoFe alloy with excellent saturation characteristics. At a gap of 3 mm the magnetic fields of the undulators were tuned by means of a Hall probe with 1 mm diameter. A magnetic field $B_0 = 0.97$ T was measured corresponding to an undulator parameter $K = 1.1$. The dipole fields were minimized by a measurement of the second field integral with a pulsed wire method [17]. Combining both measurements, $\Delta K = 10^{-3}$ and $\Delta \theta = 7.5$ $\mu$rad were reached which are well below the limits $\Delta K = 0.05$ and $\Delta \theta = 80$ $\mu$rad, at $\gamma = 1526.4$, as derived above. Details of this setup will be published elsewhere [18]. As a spectrometer a simple aluminum coated replica grating $(3 \times 3$ cm$^2$) was used with 110 lines/mm, and a blaze angle of 0.8°. With apertures of 360 $\mu$m diameter at the detector a resolving power of about 300 (FWHM) was obtained at 300 eV photon energy in the second (negative) diffraction order, satisfying fully the requirements discussed above. The resolution of the Ge$(i)$ detector of 120 eV allowed a separation of higher diffraction orders.

Typical intensity oscillations are shown in Fig. 2. The coherence without a foil is close to the maximum value $C = 1$ and does not decrease with increasing $d$. This observation is in accord with the small coherence losses expected from the discussion above. At any selected photon energy two reference measurements were performed without the foil in order to keep systematic errors small. Phase shift and amplitude were obtained from these measurements by a best-fit procedure with the function of Eq. (1). The accuracy is currently limited by systematical errors of $\pm 15$ mrad for the phase and of $\pm 2\%$ for the amplitude. The reasons for these limits are probably small horizontal and vertical drifts of the electron beam. The photon energy was determined from the period length $\lambda_d = 4\pi \gamma^2 c/\omega$ of the oscillation with an accuracy of $\pm 0.04$ eV.

The dispersion $\delta(\omega)$ and absorption $\beta(\omega)$ can only be specified if the thicknesses $t_0$ of the foils are known.
These were obtained from a phase shift measurement at the third harmonic at 897.3 eV for which \( f_1(\omega) = f_0(0) + f'(\omega) \) was taken from tables [13]. With \( f_1(H) = 1.0, f_1(C) = 6.35, f_1(N) = 8.06, f_1(O) = 7.32 \) and the stoichiometric proportions of polyimide \( H:C:N:O = 10; 22:2:5, \rho t_0 = (65 \pm 2) \mu \text{g/cm}^2 \) and \((35 \pm 2) \mu \text{g/cm}^2 \) were deduced for the carbon foil and the polyimide foil, respectively. For the densities, \( \rho = 2.26 \text{ g/cm}^2 \) and \( \rho = 1.43 \text{ g/cm}^2 \) were taken for graphite and polyimide, respectively. The results of the interference experiments are shown in Fig. 3. Typical fine structures can be recognized in the spectra which are well known from near-edge x-ray-absorption fine structure measurements [19]: the \( \pi^+ \) resonance at 285 eV, the \( C-H \) resonance at 287.5 eV, and the transition to the \( \sigma^+ \) antibonding band at 293 eV. These fine structures are observed for the first time also in the energy spectra of the real part \( \delta(\omega) \).

In conclusion, a novel interferometer has been developed. Intensity oscillations with a large degree of coherence have been observed, not only at the \( K \)-absorption edge of carbon but also at the \( L_{111} \)-absorption edge of Ni at 855 eV (at third harmonic). The energy band between 50 eV and about 1500 eV available with the interferometer at MAMI allows the investigation of many elements throughout the periodic table with samples of masses as low as 10 ng. Also, informations about the foil roughness can be obtained [18]. Furthermore, the interferometer might be well suited to solve the phase problem for thin crystals, and to study the small angle scattering of relativistic electrons in very thin foils. It may also serve as a diagnostic tool for high quality electron beams. The interferometer can be developed further into a Fourier transform spectrometer. A resolving power of 25,000 seems to be feasible if the energy spread of the electron beam can be reduced to \( 2 \times 10^{-5} \), a figure which is achievable at MAMI. The large phase shift \( \Phi_\Delta(0, d) = 10^3 \) required can be accomplished by a magnetic chicane between the undulators without loss of coherence \( C_\rho \) due to beam emittance since \( d \) remains constant. With such a chicane, also the unwanted overlap of the electron beam with the photon spot at the foil can be avoided. If the two undulators are positioned with the magnetic fields perpendicular to each other, radiation with a high degree of circular polarization can be produced [20], and the circular birefringence and dichroism of magnetic foils between the undulators can be studied.

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