Derivation of the Optical Constants of Gold from Transmission Diffraction Measurements in the 280–640-eV Range

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Measurements of the real and imaginary parts of the complex index of refraction $\hat{n}$ [$\hat{n}=(1-\delta)+ik$] of gold have been performed in the 280- to 640-eV range by a novel technique based on diffraction in transmission gratings. Uncertainties of approximately $\pm 8\%$ for δ and approximately $\pm 18\%$ for k have been attained in this first experimental determination. A theoretically predicted absorption resonance at 600 eV has been verified for the first time, and a significant disagreement with previously reported values of δ, derived from Kramers-Kronig analysis, has been established.

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Knowledge about the electromagnetic response functions (optical constants) of materials is of fundamental importance in many areas of science and engineering. There exist a large number of experimental techniques to determine the optical constants with high accuracy in certain limited spectral regions. However, our knowledge about the optical constants in the far vacuum ultraviolet (VUV) and soft x-ray regions is severely limited by the lack of accurate and direct methods of determination. In this Letter we present the first results for gold based on a novel experimental technique. This novel approach shows great promise for extension to a wide variety of materials and beyond the presently covered spectral region, 280–640 eV (44–19 Å).

The possibility of using measurements of the far-field diffraction pattern of a transmission grating to deduce the optical constants of the grating material has been previously discussed in the literature.[1,2] In a free-standing grating of rectangular bars, both the open apertures and translucent bars generate diffraction patterns in the far field and, since the diffraction pattern of the bars is both attenuated and phase shifted with respect to the aperture pattern, the two will interfere in a manner uniquely determined by the real and imaginary parts of the bars' complex index of refraction $\hat{n}$ [$\hat{n}=(1-\delta)+ik$]. To show this, consider the electric field distribution $E(x/\lambda)$ set up across the top face of the N-bar grating shown in Fig. 1 by monochromatic radiation of wavelength λ,

$$E\left(\frac{x}{\lambda}\right)=\left[M\left(\frac{x}{\lambda}\right)*\frac{1}{a}\sum_{j=-N/2}^{N/2-1}\delta\left[\frac{1}{a}\left(\frac{x}{\lambda}+\frac{d+a}{2}+ja\right)\right]\right]+\left]A\left(\frac{x}{\lambda}\right)*\frac{1}{a}\sum_{j=-N/2}^{N/2-1}\delta\left[\frac{1}{a}\left(\frac{x}{\lambda}+\frac{d}{2}+ja\right)\right]\right]. \tag{1}$$

Here, $M(x/\lambda)$ is the field distribution across a top bar surface and $A(x/\lambda)$ is the field distribution across an aperture; the asterisk denotes convolution. The intensity spectrum for the various orders m in the far field is then just the absolute value squared of the Fourier transform of $E(x/\lambda)$:

$$I^{(m)}=\left(\frac{\sin(\pi mN)}{\sin(\pi m)}\right)^2\left|\overline{M}+e^{\pi im}\overline{A}\right|^2,\quad m=0,\ \pm1,\ \pm2,\ \ldots, \tag{2}$$

where

$$\overline{M}=(a-d)\,\mathrm{sinc}[(a-d)(m/a)]e^{-2\pi iW(\delta-ik)} \tag{3}$$

with W the thickness of the bars (see Fig. 1) and $\mathrm{sinc}x=\sin(\pi x)/\pi x$, and

$$\overline{A}=d\,\mathrm{sinc}d(m/a). \tag{4}$$

In (3), the bar-vacuum reflection and phase-shift effects have been excluded, being negligible for most metals (especially Au) in the VUV and soft x-ray ranges.[1]

When we combine (2)–(4), the expression for the ratio of the mth order peak power to the zeroth-order peak power becomes

$$\frac{I^{(m)}}{I^{(0)}}=\frac{[1-2e^{-2\pi Wk(\lambda)}\cos 2\pi W\delta(\lambda)+e^{-4\pi Wk(\lambda)}]\,\mathrm{sinc}^2(md/a)}{[1+2(a/d-1)e^{-2\pi Wk(\lambda)}\cos 2\pi W\delta(\lambda)+(a/d-1)^2e^{-4\pi Wk(\lambda)}]};\quad m\neq 0. \tag{5}$$

This expression shows that, for the assumed rectangular bar profile, the ratios among all orders other than the zeroth will be *independent* of $k(\lambda)$ and $\delta(\lambda)$, and that the entire effect of $k(\lambda)$ and $\delta(\lambda)$ can be expressed by the ratio $I^{(1)}/I^{(0)}$. The establishment of the experimental equations based on (5) is immediate. If we take two gratings of the same material and of parameters a_1, d_1, W_1 and a_2, d_2, W_2 and denote the corresponding intensity ratios by

$$I_1^{(1)}/I_1^{(0)}=A, \tag{6}$$

$$I_2^{(1)}/I_2^{(0)}=B, \tag{7}$$

we get

$$\cos 2\pi W_1\delta(\lambda)-\frac{1}{1+A_1}\cosh 2\pi W_1k(\lambda)+\frac{A_1}{1+A_1}\cosh[2\pi W_1k(\lambda)+r_1]=0 \tag{8}$$

and

$$\cos 2\pi W_2\delta(\lambda)-\frac{1}{1+B_1}\cosh 2\pi W_2k(\lambda)+\frac{B_1}{1+B_1}\cosh[2\pi W_2k(\lambda)+r_2]=0, \tag{9}$$

where

$$A_1=A\times\tfrac{1}{4}\pi^2[\sin(\pi d_1/a_1)\cosh(\tfrac{1}{2}r_1)]^{-2}, \tag{10}$$

$$B_1=B\times\tfrac{1}{4}\pi^2[\sin(\pi d_2/a_2)\cosh(\tfrac{1}{2}r_2)]^{-2}, \tag{11}$$

and

$$r_i=\ln\frac{d_i}{a_i-d_i};\quad i=1,2. \tag{12}$$

It is evident that accurate measurements of the *lengths* a_i, d_i, W_i ($i=1,2$) and the *intensities* $I_i^{(0)}$ and $I_i^{(1)}$ ($i=1,2$), in principle, constitute enough data to iterate the values of k and $|\delta|$ out of Eqs. (8) and (9). In order to establish the actual validity of (8) and (9), two additional theoretical results must be established: (1) Corrections to the equations arising from nonrectangularities in actual grating bars must be derived, and (2) the attainable precision in the derived optical constants must be shown to be adequate in terms of the attainable precision in the measurements of the diffraction intensities and the grating dimensions. The first problem has been investigated, and the magnitudes of typical corrections to Eqs. (8) and (9) for Au in the soft x-ray range have been established as perturbations on the ratios $I_i^{(1)}/I_i^{(0)}$.[1,2] The second result, best expressed in the form of sensitivities of the ratios $R_i=I_i^{(1)}/I_i^{(0)}$ to the experimental parameters [where the sensitivity of x to y is defined to be $S_y^x=\partial(\ln x)/\partial(\ln y)$], is directly derivable from Eqs. (8) and (9). For example, the sensitivities of R_i to δ and k are

$$S_\delta^{R_i}=\left(\frac{2\pi W_i\delta\sin 2\pi W_i\delta}{\cosh 2\pi W_ik-\cos 2\pi W_i\delta}\right)\left(\frac{\cosh 2\pi W_ik+\cosh(2\pi W_ik+r_i)}{\cosh(2\pi W_ik+r_i)+\cos 2\pi W_i\delta}\right), \tag{13}$$

$$S_k^{R_i}=\left(\frac{2\pi W_ik\sinh 2\pi W_ik}{\cosh 2\pi W_ik-\cos 2\pi W_i\delta}\right)\left(\frac{2\cos 2\pi W_i\delta+\cosh(2\pi W_ik+r_i)-\cosh 2\delta W_ik}{\cosh(2\pi W_ik+r_i)+\cos 2\pi W_i\delta}\right). \tag{14}$$

In the experimental scheme, the gold test gratings were irradiated by filtered light emerging out of the "New Grasshopper"[3] monochromator at the Stanford Synchrotron Radiation Laboratory, and the diffraction patterns were measured 13 ft downstream by a double channel-plate detector with a 0.1-mm slit. The detector was designed to accumulate a total of 10 000 counts for each datum point in the diffracted intensity peaks and a total of 300 counts per datum point between the peaks, resulting in uniform counting statistics over the peak contours. The transmission gratings were fabricated at IBM, Yorktown Heights, with parameters particularly suitable for the experiment. The relevant dimensions of the gratings which were used to determine the constants are given in Table I. This table gives the complete set of dimensions utilized in computing the

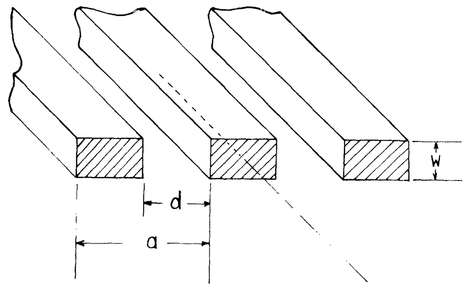

FIG. 1. Schematic of the physical structure and parameters of an ideal rectangular grating.

optical constants of Au in the spectral range 280–640 eV.

There were several sources of systematic errors in this first version of the transmission-grating diffraction experiment. The principal ones may be listed as follows.

(1) Lack of repeatability of the diffracted intensity from rotation to rotation of the sample wheel. This was caused partly by a damaged bushing on the sample wheel[2] and partly by the gratings which suffered from nonuniform parameters over their faces.[2]

(2) Normalization errors arising from both abrupt and gradual changes in the synchrotron light which appeared differently at the diffraction detector and differently at the normalization detector.[2]

(3) Asymmetry errors arising from uneven pickup of the minus-first and plus-first orders by the finite width detector slit.[2]

(4) Computational and noise errors arising from numerical processing errors, harmonics in the monochromator spectrum, and the background noise count of the detector.[2]

(5) Nonrectangular edge effects of the grating bars.[2]

With all the above errors incorporated (together with corrections arising from random surface imperfections in the grating bars[1,2]), the maximum uncertainties for the measured $I_i^{(1)}/I_i^{(0)}$ ratios were estimated to be approximately $\pm 6\%$.

The computation of the constants proceeded in two steps. First, measurements of $I_i^{(1)}$ and $I_i^{(0)}$ in the 120- and 240-eV range were used to establish values of the parameters W, a, and d for each grating (see Table I). For this computation, previously reported values of $\hat{n}$ [4] were used which had been established as reliable model predictors in earlier modeling and experimental measurements on gold transmission gratings by Delvaille *et al.*[5]

Next, by use of the values in Table I and Eqs. (6)–(12), δ and k were extracted from each data set

TABLE I. Grating dimensions used in computing the optical constants of gold.

Grating	W (Å)	d/a	Period (μm)
K12A	969.5 ± 3%	0.3117 ± 0.5%	2.5
13F	1774.6 ± 3%	0.5652 ± 0.5%	2.5
K4	1066.9 ± 3%	0.35 ± 0.5%	2.5

of $I^{(1)}/I^{(0)}$ ratios associated with each pair of gratings [(K12A,13F), (K12A,K4), (13F,K4)] and the mean values of each constant established by weighting each of the three initially computed constants with its corresponding sensitivity [see Eqs. (13) and (14)]. This process was iterated until the sensitivity values became self-consistent. (A typical sensitivity plot is shown in Fig. 2.) The estimated systematic errors and the uncertainties in the grating dimensions were also weighted by the appropriate sensitivities. The final averaged values of the constants and their uncertainties are listed in Table II. In view of the fact that the experimental measurements were dominated by systematic errors, the uncertainties shown are maximal, i.e., they represent approximately 95% confidence intervals.

The results listed represent direct measurements of the optical constants δ and k. It is important to note that all alternative metrological techniques in this energy range directly measure the absorption constants only and subsequently utilize the Kramers-Kronig (KK) relations[6] to compute $\delta(\lambda)$

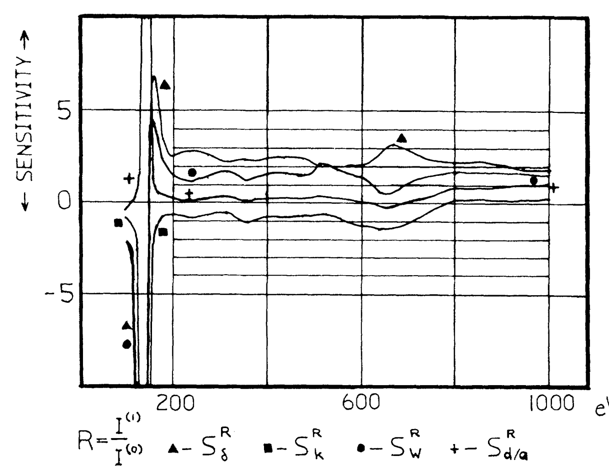

FIG. 2. Sensitivities for grating K12A over the 100- and 1000-eV range. The sensitivities over the 280- and 640-eV range are consistent with the constants tabulated in Table II, while the remaining values are taken from Ref. 4.

TABLE II. Experimental constants of gold with maximal error bounds. The approximate values in the HGK columns (Ref. 4) are interpolated.

eV	Exptl. δ	HGK δ	Exptl. k	HGK k
280	(0.0119 ± %)[a]	0.011	(0.007 62 ± %)[a]	0.00976
320	0.0109 ±8%	⋯	0.0079 ±20%	⋯
360	0.009 59 ±9%	~0.0078	0.007 62 ±17%	~0.006 982
400	0.008 72 ±8%	⋯	0.006 72 ±19%	⋯
440	0.007 98 ±8%	⋯	0.005 74 ±18%	⋯
480	0.007 29 ±8%	⋯	0.005 06 ±18%	⋯
520	0.006 95 ±7%	~0.0048	0.004 27 ±18%	~0.004 48
560	0.006 48 ±8%	⋯	0.003 78 ±17%	⋯
600	0.005 96 ±8%	0.004	0.003 29 ±15%	0.0034
640	0.005 ±13%	⋯	0.004 63 ±16%	⋯

[a]The error bounds at 280 eV are unreliable, as only one pair of gratings (K12A,K4) yielded consistent values at that energy.

from $k(\lambda)$. In view of this, it is interesting to compare our measurements with previously reported results. In Table II, values of δ and k reported by Hagemann, Gudat, and Kunz (HGK)[4] are listed. It is seen that large discrepancies exist between our measured and HGK's computed values of δ. In a more recent compilation, Henke *et al.*[7] also computes $\delta(\lambda)$ significantly different from HGK, although both his and HGK's $k(\lambda)$ values are similar. Many plausible explanations can be offered for these discrepancies, ranging from the sparseness of the $k(\lambda)$ measurements [the KK relations are exact only for continuous functions $k(\lambda)$ and $\delta(\lambda)$] to the correctness of the additive asymptotic terms in the KK relations, which depend only on theoretical assumptions and not on measurements. In the quoted analyses, however, no satisfactory resolution of the existing disagreements exists at present. A direct measurement of the kind described in this Letter is therefore extremely important, since it provides more reliable optical constants over a larger spectral region than the existing computational approaches.

As a final point, it should be noted that our measured $k(\lambda)$ displays two anomalies in the 300- and 600-eV vicinities (± 20 eV) and that neither of these has been reported earlier either by HGK[4] or Henke *et al.*[7] In this case, there is strong support for these anomalies in the recent theoretical atomic absorption cross section compilations of Plechaty, Cullen, and Howerton,[8] which indicate strong resonances both at ~ 320 and 600 eV in the total absorption cross section of Au, the latter resonance being clearly associated with the N absorption edge. The existence of these resonances also tends to qualitatively support our relatively high measured values for δ, since it is well known from dispersion analysis that a rapidly varying positive slope of k vs λ makes sizable contributions to the phase constant δ. Exact quantitative corroboration is not possible at this time, since our measurements were too sparse (± 20-eV resolution) to measure the slope of k vs λ with any reasonable accuracy. In view of these results and in view of the fact that the systematic errors in this first version of the transmission diffraction experiment can also be significantly minimized in future work, it is clear that this novel technique offers a promising approach to the future measurement of optical constants in the soft x-ray range.

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