and with  $|\Delta \delta L_A|$  = 12 ± 1  $\mu$ m,

$$
(\Delta \delta L / \Delta T)_a = (0.79 \pm 0.11) \times 10^{-2} \mu \text{m/K},
$$

 $(\Delta \delta L / \Delta T)_b = (0.13 \pm 0.02) \times 10^{-2} \mu \text{m/K}$ 

resulting from the irradiation and the reference experiments, we find  $\langle \beta_d/\beta_o \rangle$  = 12 ± 4.

A second experiment has been done recently with another test sample of zone defined aluminum. In this case two reference experiments were run, one before irradiation, one after irradiation, both yielding the same result. It led to  $\beta_d/\beta_0 = 14$ , a value consistent with the previous one.

In conclusion we have shown that in aluminum defects such as isolated vancancies and small interstitial clusters have an intrinsic thermal expansion larger by about one order of magnitude than that of the matrix  $(\beta_d/\beta_0 = 12 \pm 4)$ .

We express our gratitude to the Centre d'Etudes de Chimie Métallurgique, Vitry, which supplied zone-refined aluminum and to H. M. Gilder for several fruitful discussions.

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Impedance-Matched Grazing-Incidence Films: Pure Nuclear Reflections, Resonant Filtering of Synchrotron Radiation, and X-Ray Interferometry

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The purpose of the Letter is to point out the possibility of a new interference technique in x-ray and  $\gamma$ -ray optics—the simple idea of impedance-matched grazing-incidence films  $-$ and to discuss applications to pure nuclear coherent reflections, resonant filtering of synchrotron radiation, and x-ray interferometry.

In x-ray optics, the small index-of-refraction change  $(N_0 = 1 - N_1 = 1 - 2\pi\lambda^2 n z r_0 \approx 1 - 10^{-5}$ , where  $n$  is the atomic density,  $z$  is the atomic number, and  $r_0 = e^2/mc^2$  on entering a medium of higher electron density produces near total reflection of x rays near grazing incidence on a flat surface, with the critical angle being  $\varphi_c = (n\lambda^2 z r_0/\pi)^{1/2}$ . For  $\varphi > \varphi_c$ , there is a rapid dropoff in reflected intensity with increasing  $\varphi$ , and a rapid increase in the penetration depth  $l_{\perp}(\varphi)$ . For Fe<sup>56</sup>, and 14.4-keV radiation,  $\varphi_c = 3.8$  mrad, and for  $\varphi = 3.0$ mrad, then  $|R|^2 \approx 0.9$  and  $l_+ \approx 29$  Å, while at  $\varphi$ =4.5 mrad, then  $|R|^2 \approx 0.1$  and  $l_1 \approx 472$  Å.

For  $\varphi > \varphi_c$ , where deep penetration occurs, it should be possible to strongly suppress the x-ray reflection with an impedance-matched quarterwave film, just as in optical coating of lenses, That is, the mirror is coated with a film of proper impedance such that the reflections at the upper and lower interfaces are equal and with the thickness chosen so the two waves emerge 180' out of phase.

The quarter-wave-plate condition gives  $l_F(\varphi)$  $\lambda(\varphi^2 - \varphi_c^2)^{-1/2}$ , which is typically  $\approx 50-100$  Å. A quarter-wave plate for  $1-\AA$  radiation is only possible because grazing incidence augments the

636 1979 The American Physical Society

required film thickness by the factor  $(\varphi^2 - \varphi_c^2)^{-1/2}$ .

Explicitly, for grazing-incidence reflection from a mirror  $(2)$  coated with a film  $(1)$  of thickness  $l_F$ , the reflection amplitude is<sup>1</sup>

$$
R(\varphi) = \frac{R_{01} + R_{12} \exp(i2g_1 l_F)}{1 + R_{01} R_{12} \exp(i2g_1 l_F)},
$$
\n(1)

where  $R_{01}$  and  $R_{12}$  are the reflection amplitudes at the medium interfaces  $0-1$  and  $1-2$ , respectively,  $R_{01}(\varphi) = (1 - \beta_1)$ , and  $R_{12}(\varphi) = (\beta_1 - \beta_2) / (\beta_1)$ + $\beta_2$ ), the complex wave vector in film 1 is  $g_1$  $=k_0 \varphi \beta_1$ , and, for medium j,

$$
\beta_1 = \{ 1 + [n(j)\lambda^2 f_e(j)/\pi \varphi^2 ] \}^{1/2}
$$
  
=  $\{ 1 - (\varphi_c(j)/\varphi)^2 + i [\pi \varphi_e(j)/\varphi^2] \}^{1/2}$ . (2)

Here  $f_e$  is the atomic forward-scattering amplitude due to electronic scattering,  $f_e = -(z + \Delta f')r_0$ + $i(\sigma_e/4\pi\lambda)$ , when  $zr_0$  is the Thomson scattering, is the anomalous scattering (only apprecia ble near an absorption edge), and  $\sigma_e$  is the absorption cross section. The critical reflection angle is  $\varphi_c = [n \lambda^2 (z + \Delta f') r_0 / \pi]^{1/2}$ .

The impedance-match condition  $R_{01} = R_{12}$  reduces to  $\beta_1^2 = \beta_2$ , which requires an electron density

$$
\rho_1(\varphi) \equiv n_1(z_1 + \Delta f_1')
$$
  
\n
$$
\approx (\pi \varphi^2 / \lambda^2 r_0) (1 - \{1 - [\varphi_c(2) / \varphi]^2\}^{1/2}), \quad (3)
$$

and for a quarter-wave plate, the film thickness should be an odd multiple of

$$
l_F(\varphi) \approx \left\{ \frac{1}{4} \lambda \left[ \varphi^2 - \varphi_c(1)^2 \right]^{1/2} \right\}.
$$
 (4)

Here we have neglected photoabsorption. For a coating of given  $\rho_1$ , Eq. (3) determines the angle  $\varphi_0$  at which an impedance match occurs, and for a quarter-wave plate, the thickness should be taken as  $l_F(\varphi_0)$ .

Both the required electron density and proper



FIG. 1.  $|R|^2$  vs  $\varphi$  for Fe<sup>56</sup> coated with: (1) uncoated (2) 70 Å Te, (3) 75 Å Te, (4) 80 Å Te, and (5) 85 Å Te.

film thickness depend on the angle of incidence  $\varphi$ . An examination of Eq. (3) shows that the required electron density varies from  $\rho_1(\varphi) \approx \frac{1}{2} \rho_2$ for  $\varphi \gg \varphi_c(2)$ , increasing to  $\rho_1(\varphi) \approx \rho_2$  for ing  $\varphi \rightarrow \varphi_c(2)$ , and the required  $l_F$  increases with decreasing  $\varphi$ .

For an Fe<sup>56</sup> mirror  $(\rho_2 = 2.24 \text{ Å}^{-3})$ , there are many possible coatings. As an example we take Te( $\rho_1 = 1.53 \text{ Å}^{-3}$ ). Figure 1 shows rocking curves  $|R|^2$  vs  $\varphi$  for different thicknesses of Te. We note the position  $\varphi_{\min}$  of minimum reflectivity changes with thickness and there is an optima thickness  $l_F \approx 80$  Å and optimal angle of incidence  $\varphi_{\min} \approx 4.2$  mrad for maximum suppression. This qualitatively agrees with the approximations in Eqs. (3) and (4), but the quantitative results are shifted significantly by the effects of photoabsorption.

Three main factors limit the degree of suppression: photoabsorption; incident beam divergence; and film thickness nonuniformity. tion gives an intrinsic limit to suppression by introducing a complex component into the reflectivities which prevents exact impedance matching. Beam divergence is a limiting factor due to the  $\,$ rapid variation of  $|R(\varphi)|^2$  vs  $\varphi$ , and similarly film thickness variations change  $\varphi_{\text{min}}$ . Thus it's necessary to average over the angular divergence of the beam and film thickness variations.

For DORIS, the divergence of the x-ray beam at 10 keV is about 0.25 mrad. Thicknes in producing thin films is about  $\pm 2.5\%$  absolut thickness.

Table I gives the numerically determined optimal  $l_F$ ,  $\varphi$ <sub>min</sub>, and the average reflectivities for several films on Fe, averaged over a beam divergence  $\varphi_0 \pm 0.13$  mrad, and over thickness varia tions  $l_F \pm .025l_F$ . We see that strong suppressions still occur, with reduction factors  $|R|^2/|R|_{Fe}^2$  $\approx 10^{-3} - 10^{-2}$ .

Of course a crucial question in these considera

TABLE I. Optical parameters  $l_F$ ,  $\varphi_{\text{min}}$  and minimum reflectivities for several coatings on Fe reflecting 14.4 keV radiation.

EL	$\mathbf{\hat{A}}$ $-3$	Φ $(10^{-3})$ rad)	$l_F$ A)	$ R ^2$ $(10^{-4})$	$\overline{\mathbf{2}}$ $ R _{Fe}$ $R$ <sup>2</sup>
Ti	1.24	6.0	42	0.24	719
Se	1.25	6.0	40	0.21	807
Ge	1.42	4.6	65	3.7	245
Te	1.53	4.4	76	9.4	135

tions is what flatness is required to be able to achieve effective quarter-wave films? High-quality optically flat plates have a long-range flatness typically  $\approx \lambda/20$ , but microscopically the surfaces will resemble a rippling flag rather than the idealized flat surface. However, critical reflection experiments indicate that perfect flatness to an angstrom scale is not necessary. For critical reflection of x rays from optically flat plates penetration depths are only  $\approx 20$  Å, yet the reflectivities are very close to the theoretical predictions, and there is no significant increase in the beam divergence as would be expected if the scattering was sensitive to surface irregularities. In effect, the coherent scattering "sees" the average flatness and not the detailed graininess. Qualitatively, coherent scattering from an irregular surface can be viewed as reflection from the average flat surface, plus diffuse scattering from the deviations from flatness which tends to cancel out. If this view is correct, then optically flat plates should suffice for quarter-wave films. However, if detailed surface graininess is important, quarter-wave films may still be possible since very high-quality optically flat plates are now available with a long-range flatness of  $\lambda$  /50 and with a microscopic graininess of  $\leq 5$  Å.

Finally we note that electronic scattering can also be suppressed with a half-wave film: For a (hypothetical) free film the negative impedancematch condition  $R_{12} = -R_{01}$  holds for any angle, and suppression of the electronic scattering will then occur for a half-wave condition,  $\exp(i2g_1'l_F^{\top})$ = +1 or  $l_F(\varphi) \approx \frac{1}{2} \lambda (\varphi^2 - {\varphi_c}^2)^{-1/2}$ . The films must of course, be on an optically flat backing, which will produce a small uncanceled reflection, and so the degree of suppression for  $\lambda/2$  films is not as strong as for impedance-matched  $\lambda/4$  films. but  $\lambda/2$  films do offer the advantage that they can be operated at any desired angle by appropriate choice of film thickness.

Although it is not possible to go into detail here, we now very briefly discuss three applications of impedance-matched grazing-incidence films. Details are given elsewhere. '

Pure nuclear reflections.—The interesting phenomena of coherent scattering of  $\gamma$  rays by nuclei has received extensive theoretical and experimental effort from the very early days of the Mössbauer effect. The first theoretical treatment of  $\gamma$  optics was given by Trammell<sup>2</sup> in 1960. and concurrently the first dynamical experiments were performed by Black and Moon' on Bragg reflection, and by Bernstein and Campbell' on grazing-incidence reflection. Further theoretical developments were given by Afanas'ev and Kagan' in 1965.

Particular attention has been given to methods of suppressing the electronic scattering to obtain very pure nuclear reflections, and several Bragg reflection techniques have been found: antiferromagnetic and other superlattice reflections, 90 Bragg reflection of polarized radiation, and crystalline thin-film reflections.

The alternative proposed here is grazing-incidence reflection from a resonant mirror coated with an impedance-matched quarter-wave film. The essential point is that while off resonance the reflection is strongly suppressed, for nearresonance radiation the index of refraction in the resonant medium is strongly altered giving an impedance mismatch and strong reflection.

For a resonant medium covered with a quarterwave film, the reflection amplitude is again given by Eq. (1), but  $\beta_2$  now includes the nuclear scattering,

$$
\beta_2 = \left\{ 1 - \left[ \varphi_c(2)/\varphi \right]^2 + in \lambda \sigma_e / \varphi^2 + n \lambda^2 \mathfrak{F}_N / \pi \varphi^2 \right\} .^{1/2}
$$

Here  $\mathfrak{F}_{N}$  is the nuclear forward-scattering amplitude,  $\mathfrak{F}_{N} = \sum \mathfrak{F}_{0}(m_{0}, m_{1})/[x(m_{0}, m_{1}) - i]$ , where  $\mathfrak{F}_{0}(m_{0},m_{1})$  is the oscillator strength for the  $j_{0},m_{0}$  $j_1, m_1$  transition, and  $x(m_0, m_1) = 2[E(j_1, m_1)]$  $-E(j_0, m_0) - \hbar \omega$  /  $\Gamma$ . With a perfectly impedancematched quarter-wave film, the reflectivity becomes (neglecting photoabsorption)

$$
R(\varphi, \omega) = \frac{1 - \{1 + n\lambda^2 \mathfrak{F}_N / \pi [\varphi^2 - \varphi_c(2)^2] \}^{1/2}}{1 + \{1 + n\lambda^2 \mathfrak{F}_N / \pi [\varphi^2 - \varphi_c(2)^2] \}^{1/2}},
$$
(5)

explicitly demonstrating that this is a pure nuclear reflection.

In Figure 2 we plot  $|R|^2$  vs  $\omega$  for an Fe<sup>57</sup> mirror coated with a 76- $\AA$  Te film at  $\varphi = 4.4$  mrad.



FIG. 2.  $|R|^2$  vs  $\Delta$  (= $\hbar\omega - E_{res}$ ) for Fe<sup>57</sup> coated with 76 Å Te,  $\varphi = 4.4 \times 10^{-3}$ , for incident  $\hat{\epsilon}_{(+1)}$  (solid) and  $\hat{\epsilon}_{(-1)}$ (dashed) .

Here the internal  $\vec{B}$  field is aligned  $\|\vec{k}_{0} \|$  so that the eigenpolarizations are the circular polarizations  $\hat{\epsilon}_{\text{(H)}}$  which couple, respectively, to the two  $\Delta J_z = \pm 1$  transitions. The solid line gives the response for incident  $\hat{\epsilon}_{(n)}$ , the dashed for  $\hat{\epsilon}_{(n)}$ . We note in particular that very strong reflections occur, with peak reflectivities  $\simeq 0.7$  and strongly broadened widths  $\Gamma_{eff} \simeq 20$ .

The strong broadening is very important and is due to the "double enhancement effect": First a broadening  $\approx \lambda n \sigma_0 \Gamma/2\varphi^2 \approx 5\Gamma$  due to the "enhanceof badening  $\sim$   $n\sigma_0$   $\sim$   $/2\gamma \approx 51$  due to the emandement effect"<sup>2</sup> (for waves incident near Bragg or grazing incidence on a resonant medium, there is a broadened width to the frequency response due to coherent reemission into the reflection channel, and, correspondingly, the time response is speeded up relative to the natural lifetime for incoherent decay and internal- conversion absorption), and secondly an augmentation by [1  $-(\varphi_{\rm c}/\varphi)^2]^{-1} \approx 4 \times$  due to refraction [because of refraction, the effective angle of the wave driving the resonant system is decreased to  $\varphi$ [1 –  $(\varphi$ /  $\varphi$ <sup>2</sup>],<sup>1/2</sup> which in turn augments the enhancement broadening].

Synchrotron filtering. —An important application of this technique is to Mössbauer filtering of synchrotron radiation. Here we summarize the main conclusions.

(1) Effective filtering can be achieved in 2-4 multiple reflections depending on the quarterwave film used. For a fourfold reflection from  $Fe<sup>57</sup>$  mirrors coated with Te, the signal to noise ratio (number of resonant photons  $N_{res}/$ number of nonresonant photons  $N_{\text{nonres}}$  in a 10-eV width) is  $\simeq$  1.3  $\times$  10<sup>3</sup>!

(2) Because of double enhancement, and the strong reflectivities,  $N_{\text{res}}$  will exceed  $\Gamma I_0 \Delta_x \Delta_y$ , the quanta incident within a natural  $\Gamma$ .  $(I_0 = \text{inci}$ dent photons/eV/mrad<sup>2</sup>;  $\Delta_x, \Delta_y$  give the vertical and horizontal divergence of the beam). For the fourfold reflection with Te films  $N_{res} \simeq 2.1 \Gamma I_0 \Delta_x \Delta_y$ .

(3) In comparison to Bragg techniques the resulting resonant fluxes are potentially 2-3 orders of magnitude greater. Furthermore, there are very severe problems with Bragg filters, all centered on the very restrictive nature of the Bragg condition. These problems are all eliminated by the proposed technique since this is an index- of-refraction technique.

 $X$ -ray interferometry. - Impedance - matched grazing-incidence films work on interference

and are limited form of x-ray interferometer which can be applied to some of the same problems currently investigated by Bonse-Hart interferometry.

For example, an important and difficult problem is to obtain precise measurements of the anomalous scattering  $\Delta f'$  in the vicinity of an absorption edge.<sup>6</sup> An impedance-matched quarter-wave film interferometer could also be used for such studies by isolating the anomalous scattering: The impedance match can be done just to eliminate the Thomson scattering leaving a pure anomalous scattering reflection,

$$
R(\varphi) \simeq \frac{1 - [1 - n\lambda^2 \Delta f'/\pi (\varphi^2 - \varphi_c^2)]^{1/2}}{1 + [1 - n\lambda^2 \Delta f'/\pi (\varphi^2 - \varphi_c^2)]^{1/2}}.
$$
 (6)

Here we have neglected photoabsorption, which of course is important. We note that just as for a pure nuclear reflection, there is an important index-of -ref raction augmentation.

Impedance-matched film interferometry lacks the precision of Bonse-Hart interferometry, but would appear to offer a complementary technique. particularly for studies of lower-energy absorption edges  $\zeta$  1 keV where wave lengths are too long for Bragg reflections in a Bonse-Hart system.

Note added.-It has been pointed out to us by Dr. G. Materlik that half-wave-plate interference fringes for Ni on glass were observed by Kiessig' in 1930. We now have preliminary measurements of suppression by impedance-matched quarterwave films of Ge on Fe. These results will be reported later.

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