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Model Comparisons With Glancing Incidence Measurements of Overcoated Metal Mirrors

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The absorptance characteristics of metal mirrors, with thin dielectric overcoats on their surface, have been measured at angles of incidence from 0 to 89°, for both s and p-polarizations. The mirrors tested are silver overcoated with ThF_4, and aluminum overcoated with MgF_2. To understand the experimental results requires the use of models incorporating thin film theory. Estimations of parameters, such as film thickness, can be made by varying parameters within the model until the best fit with the experimental data is obtained. This paper presents the results of model comparisons with the glancing incidence data. In addition, thin film designs for overcoated mirrors with enhanced reflectivity at glancing incidence are discussed.

Key words: thin film model; glancing incidence; overcoat; optimum mirror design; silver; aluminum; ThF_4; MgF_2.

1. Introduction

Glancing or grazing incidence mirrors are critical components in some laser system designs. For example, a proposed [1] free electron laser for generation of XUV or soft x-rays uses a ring resonator comprised of many mirrors adjusted to operate at glancing incidence. Although bare metal mirrors are logical choices for glancing incidence applications, some preferred metals, such as silver and aluminum, have problems with tarnishing. It has been shown [2] that the absorptance, caused by the tarnish, tends to be worse at glancing incidence than at normal incidence. One way to prevent the tarnishing is to apply a protective dielectric overcoat on the metal surface. However, there has been little work done to understand how an overcoated metal mirror behaves at high angles of incidence.

A photoacoustic calorimetry system [3] has been developed at Spectra Technology, Inc. (STI) and has been successfully used to measure the absorptance characteristics of bare metal mirrors from 0 to 89° [4]. The system has also been used to measure the glancing incidence absorptance characteristics of overcoated metal mirrors. The results for two mirrors, MgF_2 on aluminum and ThF_4 on silver, are discussed in this paper.

While simple Fresnel theory for bare metals [5] cannot be used to predict the absorptance behavior of overcoated mirrors, the theory of thin films on surfaces is well established [6], and computer codes are available to accurately predict the optical characteristics of films on metal surfaces. This paper discusses the application of thin film theory to interpret the glancing incidence experimental data. In addition, examples are given of model predictions for thin films designed to not only overcoat the metal surface, but at the same time enhance the reflectivity at a high angle of incidence for a given polarization.

2. Review of Thin Film Theory

As mentioned earlier, there has been a great deal of work in the area of optical thin films; Macleod's [6] book is a classic reference. Hence, the description in this paper shall be restricted to the codes and basic equations used in this work.

Figure 1 is a cross section of a single layer film on a substrate. The index of refraction n_2 is assumed to equal 1 (air). The film is assumed to be non-absorbing (i.e., the extinction coefficient, k, is zero) and has a thickness d with an index n_1. This is a reasonable assumption for most films at the visible and IR wavelengths examined during this program. The absorbing substrate has a complex index of refraction given by n - ik. The Fresnel reflectance and transmittance coefficients [5] at each of the boundaries are designated by r_n and t_n, n = 1, 2, respectively. For a single layer, non-absorbing film on an absorbing substrate, the reflectance as a function of angle of incidence is given by [7],

$$R = \frac{r_2 + r_1\exp\left(-2i\delta_1\right)}{1 + r_1r_2\exp\left(-2i\delta_1\right)} \qquad [1]$$

where $\delta_1 = (2\pi n_1 d\cos\phi)/\lambda$ is the phase thickness at angle ϕ within the film.

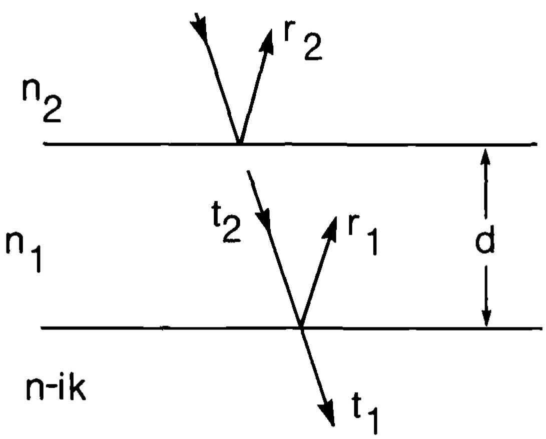


Figure 1. Schematic cross section of a non-absorbing thin film on an absorbing substrate. See text for explanation.

Equation 1 is valid only for a single layer, non-dispersive film. More complex films can be analyzed using the characteristic matrix formalism. The elements of the matrix are solutions to the wave equation, which satisfy the conditions of continuity of the tangential components of the electric and magnetic fields across the plane film boundaries. Sophisticated computer models have been developed, which are capable of analyzing multi-layer films, dispersive (absorbing) films, films with gradients, and so on. However, for the data presented in this paper, the simple expression given by Eq. (1), reveals much about the characteristics of the mirrors tested. Hence, all the theoretical curves displayed in this paper are based upon Eq. (1).

3. Comparisons of Experiment With Theory

The glancing incidence absorptance measurement system has been described in detail elsewhere [3]. It uses photoacoustic calorimetry to detect the laser energy absorbed on the mirror surface as a function of the incidence angle. Absolute absorptance is determined using a laser energy ratiometer. The uncertainty of the absolute absorptance measurement is ≈±20%; error bars on the data, given in this paper, represent the reproducibility of the photoacoustic measurements and not the variance in the absolute value of the absorptance.

Figure 2 shows the measured absorptance characteristics of an aluminum mirror with a MgF_2 overcoat. The laser wavelength for all the work discussed in this paper is 0.5145 μm (argon ion laser). Figure 2(a) is for s-polarization (perpendicular to the plane of incidence); Figure 2(b) is for p-polarization (parallel to the plane of incidence). The solid curves in Figure 2 are the Fresnel equation predictions for a bare metal surface, using the values of n (0.819) and k (6.26) given by Shiles, *et al.* [8]. Not surprising, the bare metal theoretical curves are very poor at predicting the absorptance characteristics of the overcoated aluminum mirror.

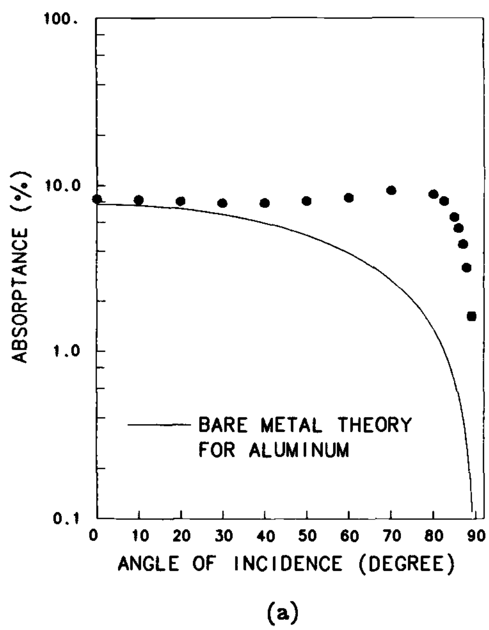
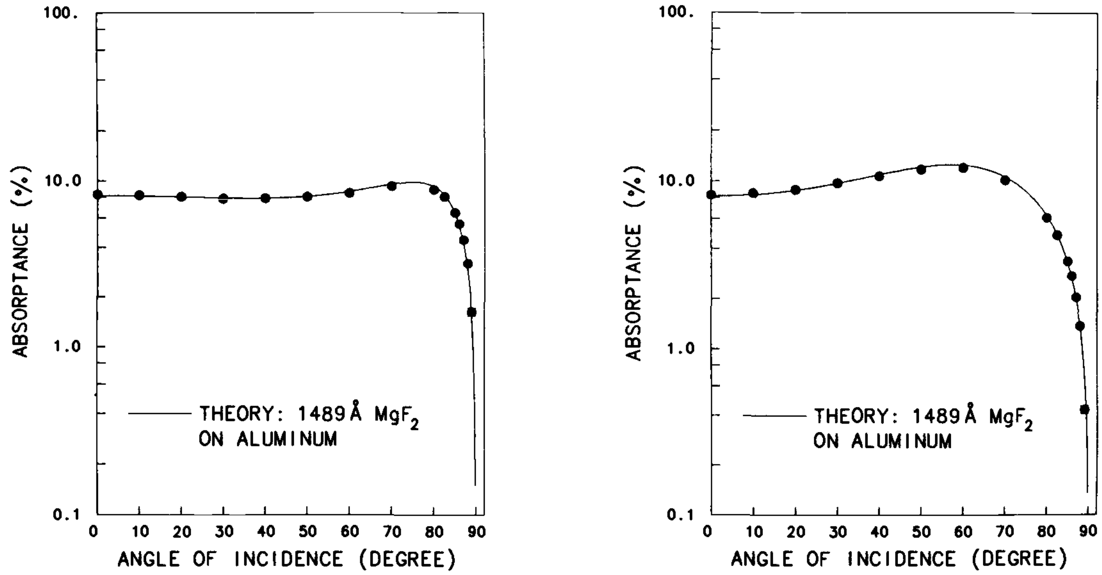


Figure 2. Measured absorptance characteristics of a MgF_2 overcoated aluminum mirror at 0.5145 μm. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the Fresnel theory prediction for a bare metal surface using the values of n and k given by Shiles, *et al.* [8].

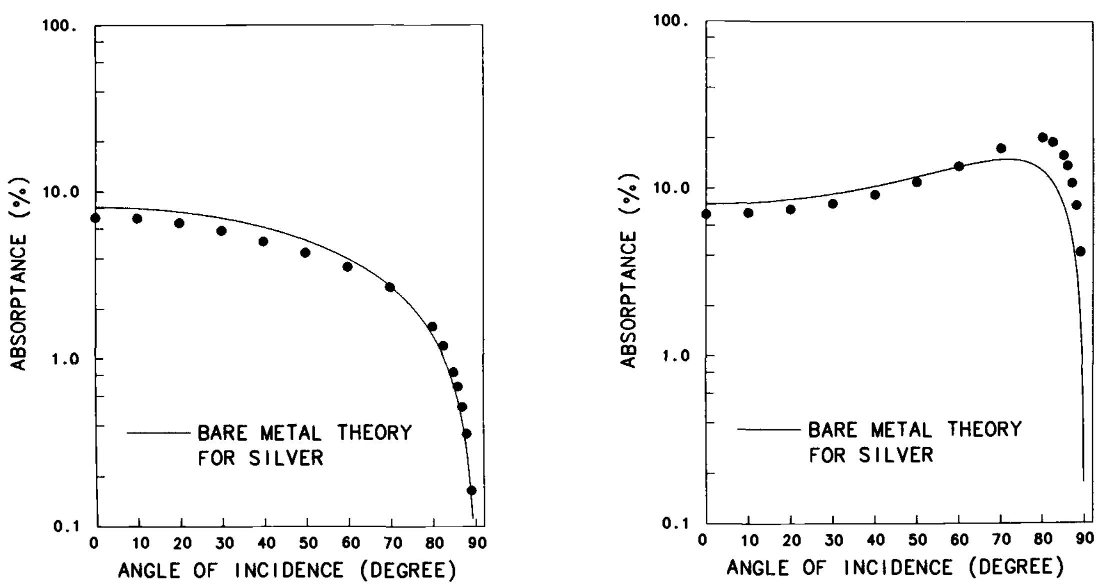
Figure 3 shows that the agreement between the data and theory is much better when using a thin film model. In this case, the computer code varied the thickness of the MgF_2 coating ($n = 1.38$ [6]), until the best fit with the data was obtained at a thickness of 1489 angstroms.



(a) (b)

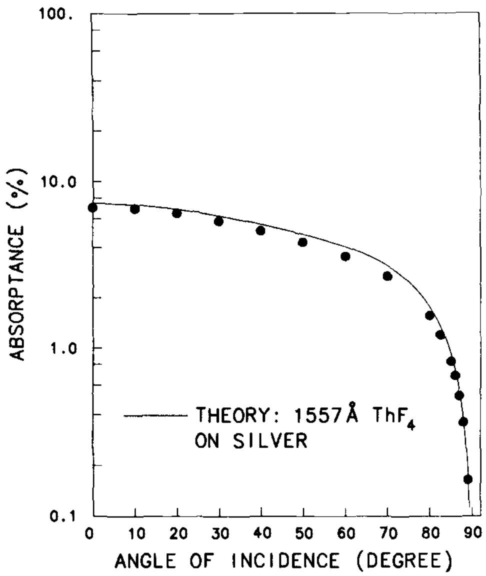
Figure 3. Optimum fit of thin film model with experimental data. The data is the same as given in Fig. 2. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction assuming a MgF_2 thickness of 1489 angstroms.

The experimental data for silver overcoated with ThF_4 is plotted in Figure 4, along with the Fresnel equation predictions for a bare metal surface using the values of n (0.245) and k (3.25) given by Hagemann, *et al.* [9]. Although, the bare metal predictions are closer in agreement in this case than for the overcoated aluminum mirror, there are still significant differences evident. Applying the thin film code again, optimum fit is obtained if a ThF_4 (n = 1.53 [6]) thickness of 1557 angstroms is assumed; this is shown in Figure 5.

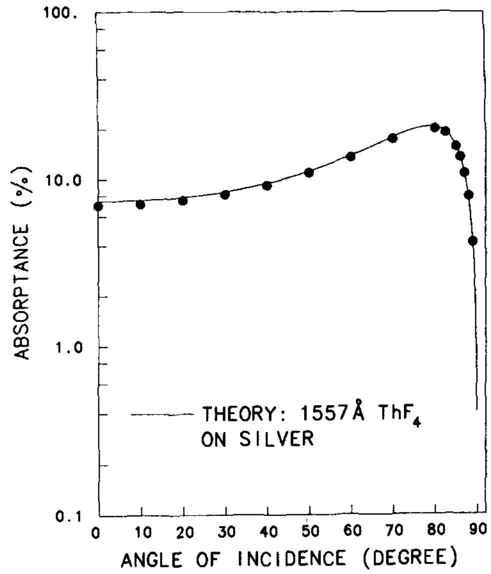


(a) (b)

Figure 4. Measured absorptance characteristics of a ThF_4 overcoated silver mirror at 0.5145 μm. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the Fresnel theory prediction for a bare metal surface using the values of n and k given by Hagemann, *et al.* [9].



(a)



(b)

Figure 5. Optimum fit of thin film model with experimental data. The data is the same as given in Fig. 4. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction assuming a ThF_4 thickness of 1557 angstroms.

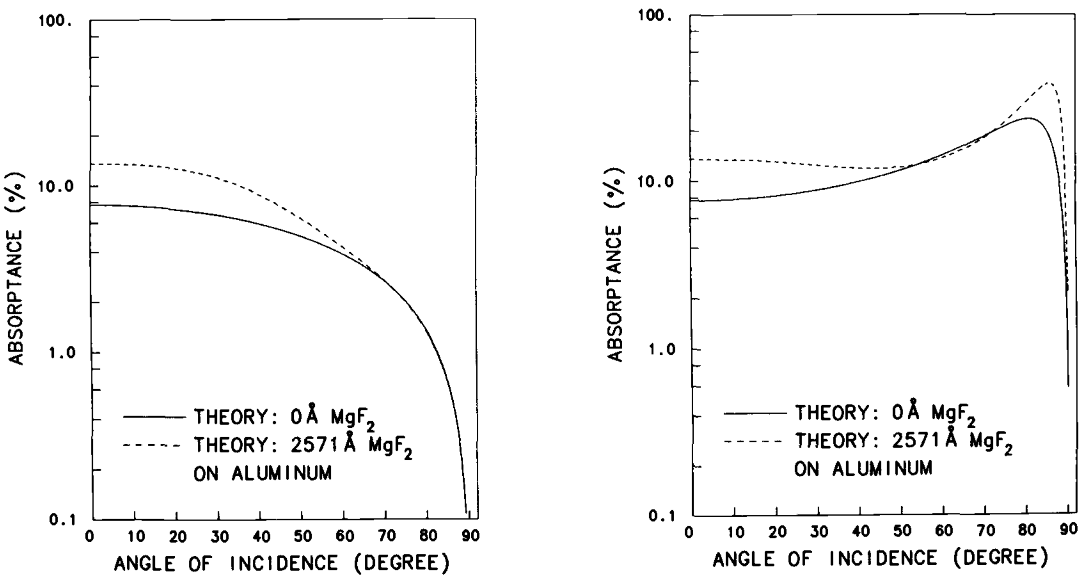
4. Discussion

Agreement between the thin film model and the experimental data is very good. The fact the agreement remains good over the full range of incidence angles and for both polarizations is a strong indication that the solution determined by the model (i.e., the film thickness for optimum fit) is a unique one. The actual thicknesses of the films on these mirrors is currently being measured by independent means.

Given the ability to predict accurately the absorptance (or reflectance) properties of overcoated mirrors at glancing incidence, it is interesting to ask the question: what film thickness will minimize the absorptance (and thereby maximize the reflectance) of an overcoated mirror at a specified polarization and high angle of incidence? From the data shown in the previous section, it is clear that arbitrarily choosing a film thickness can result in adversely affecting the absorptance of the mirror at high angles of incidence.

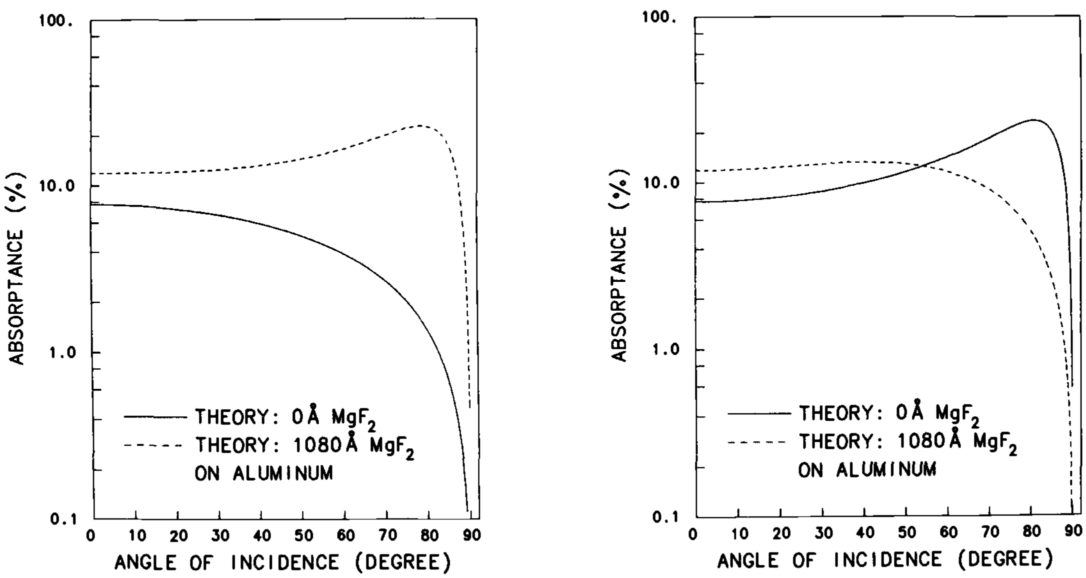
To answer this question, the MgF_2 film thickness on the aluminum mirror is systematically varied in the model until the minimum absorptance is predicted. The results for minimizing the s-polarization at 85° are shown in Figure 6. The model predicts an optimum thickness of 2571 angstroms MgF_2; for comparison the theoretical curve for bare metal (i.e., 0 angstroms MgF_2) is also plotted. At angles greater than roughly 70°, the thin film absorptance curve is very similar to the bare metal curve and, therefore, its absorptance at 85° is essentially as low as a bare metal. There is, however, a penalty associated with the film. The absorptance at normal incidence is higher than for a bare metal, and the p-polarization absorptance at glancing incidence has increased over nearly all angles.

Figure 7 shows the theoretical results for a MgF_2 film on aluminum designed to minimize the p-polarization absorptance at 85°. At a film thickness of 1080 angstroms, the p-polarization absorptance curve no longer displays a Brewster angle effect at high angles of incidence, but instead has a curve shape similar to that for s-polarized light on bare metal. Although the p-polarization absorptance has become smaller at high angles of incidence, it is not as small as the s-polarization absorptance of a bare metal. Note, in Figure 7(a), that the s-polarization absorptance now increases dramatically with angle.



(a) (b)

Figure 6. Model predictions of MgF_2 overcoated aluminum. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction for a bare metal surface; the dashed curves for a film thickness designed to minimize the s-polarization absorptance at 85°



(a) (b)

Figure 7. Model predictions of MgF_2 overcoated aluminum. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction for a bare metal surface; the dashed curves for a film thickness designed to minimize the p-polarization absorptance at 85°.

Figures 8 and 9 show the results of similar efforts to determine the thicknesses of ThF_4 films on silver, which minimize the absorptance at 85° for s and p-polarization, respectively. In Figure 8(a), with 2007 angstroms ThF_4, the absorptance of the s-polarization is actually slightly less than it is for a bare metal.

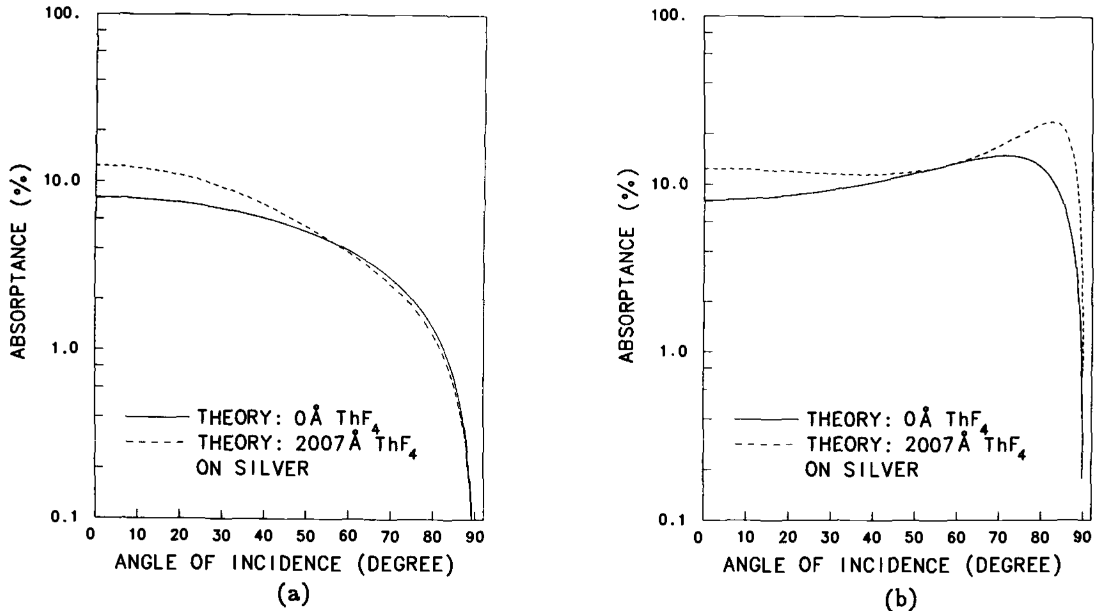


Figure 8. Model predictions of ThF_4 overcoated silver. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction for a bare metal surface; the dashed curves for a film thickness designed to minimize the s-polarization absorptance at 85°.

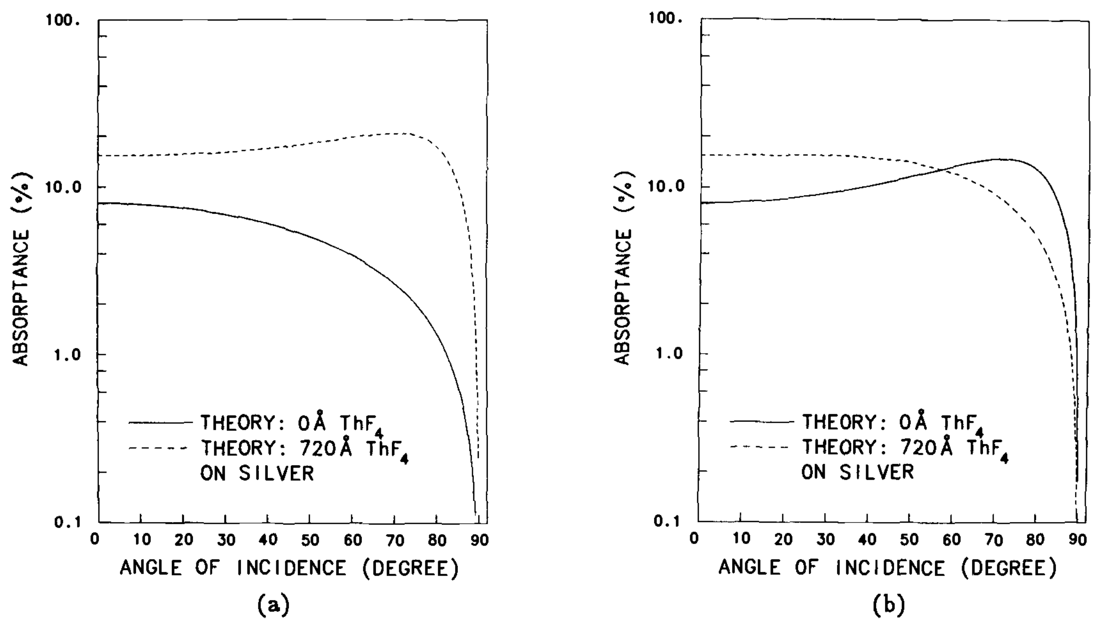
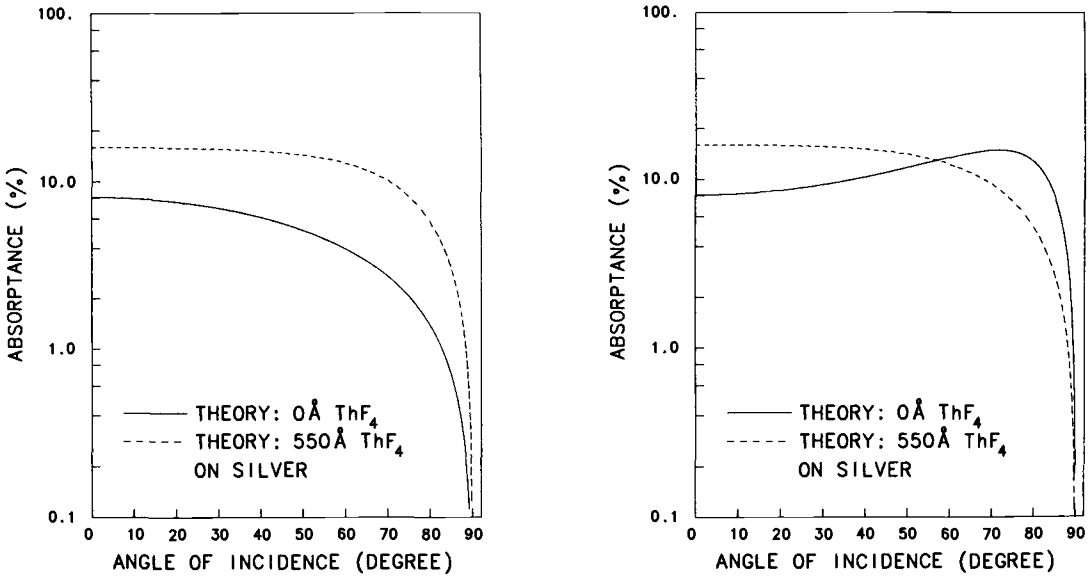


Figure 9. Model predictions of ThF_4 overcoated silver. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction for a bare metal surface; the dashed curves for a film thickness designed to minimize the p-polarization absorptance at 85°.

It is apparent when comparing Figures 6 and 7, and Figures 8 and 9, that the absorptance characteristics tend to change with film thickness in opposing manners for each polarization. This implies there exists a film thickness which causes both polarization components to display a similar angular dependence. Indeed, this is demonstrated in Figure 10, which shows that a 550 angstrom ThF_4 film on silver causes both polarizations to have nearly identical absorptance behavior. Such a film thickness may be important when high reflectivity at both polarizations is needed at high angles of incidence.



(a) (b)

Figure 10. Model predictions of ThF_4 overcoated silver. (a) is for s-polarization; (b) is for p-polarization. The solid curve is the prediction for a bare metal surface; the dashed curves for a film thickness designed to make the absorptance characteristics similar for both polarizations.

5. Conclusion

Thin film models provide an accurate means of interpreting the glancing incidence measurements of dielectric overcoated mirrors. They can also be used to predict the best film thickness to apply, in order to optimize a desired characteristic of the mirror at a high angle of incidence. This is important because of the need to apply overcoats on bare metal mirrors, such as silver, to prevent tarnishing. It may be also possible to design films that yield reflectivities at glancing incidence higher than is obtainable with a bare metal. Further study is needed to determine whether this can be obtained with a single layer dielectric on metal or would require multi-layer dielectric coatings.

6. Acknowledgments

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7. References

[1] Newnam, Brian E. Multifacet - all metal reflector design for soft x-ray and XUV free electron resonators. 17th Annual Symposium on Optical Materials for High Power Lasers, Boulder, CO, October 28-30, 1985.

[2] Kimura, Wayne D. Comparison of bare silver and bare silver on copper mirrors. Optical Society of America Workshop on Optical Fabrication and Testing, Seattle, WA, October 21-23, 1986, paper no. IV-WB8.

[3] Kimura, Wayne D.; Ford, Dennis H. Photoacoustic calorimetry system for glancing incidence mirror absorptance measurements. Rev. Sci. Instrum. 57(11): 2754-2762; 1986.

[4] Kimura, Wayne D.; Ford, Dennis H. Absorptance measurements of metal mirrors at glancing incidence. Appl. Optics. 25(20): 3740-3750; 1986.

[5] Born, Max; Wolf, Emil. Principles of optics: Pergamon Press; 1975. 628-630.

[6] Macleod, H. Angus. Thin-film optical filters: American Elsevier; 1969.

[7] Heavens, O.S. Optical constants of thin films, in Physics of thin films, vol. 2: Academic; 1964. 203.

[8] Shiles, E.; Sasaki, Taizo; Inokuti, Mitio,; Smith, D.Y. Self-consistency and sum-rule tests in the Kramers-Kronig analysis of optical data: application to aluminum. Phys. Rev. B. 22(4): 1612-1628; 1980.

[9] Hagemann, H.J.; Gudat, W.; Kunz, C. Optical constants from the far infrared to the x-ray region: Mg, Al, Cu, Ag, Au, Bi, C, and Al_2O_3. J. Opt. Soc. Am. 65(6): 742-744; 1975.