研究論文

Calculation of the Effect of Surface Roughness and Carbon Contamination on the Reflectivity of an X-ray Mirror

表面粗糙度及碳污染對 X 光鏡片反射率影響之計算

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ABSTRACT

The surface contamination and roughness on the X-ray mirror affect the ability of X-ray reflectivity, thus it reduces the X-ray flux delivered to the sample position. In this report, we calculated the effects on the reflectivity of an X-ray mirror due to the surface roughness and surface contamination, mainly, the carbon deposition on the surface of a mirror. In order to gain at least 70% of reflectivity from a Pt coating mirror (8 KeV, 6 mrad of incidence angle), the thickness of the carbon film must be kept below 4500 Å . We also estimated the surface roughness required for the Pt coating mirror. The result showed that the surface roughness should be kept below 33 Å.

摘　要

X 光鏡片運用掠角入射之幾何條件可以使 X 光於表面上產生反射進而聚焦。然而鏡片表面的碳污染或其粗糙度嚴重地影響其反射率，因此也降低了 X 光射源聚焦到試樣上光子之通率。本報告中，我們將對於 X 光鏡片表面的粗糙度及碳沈積所造成反射率降低之影響加以計算。所得之結果如下：若欲使鍍鉑之 X 光鏡片對於 8KeV 之 X 光之反射率達到 70%以上，在 6mrad 之入射角之下，對碳沈積而言，其累積之厚度必需小於 4500 埃，而對於乾淨之鍍鉑鏡片而言，其表面之粗糙度則需在 33 埃以下。

I. Introduction:

The X-ray mirrors operated under grazing incidence geometry were used to focus the soft and hard X-rays from a near collimated source, such as synchrotron radiation source or X-rays from a research X-ray generator. The quality of a mirror surface decides the ability of the X-rays reflection from a surface, which also affects the X-ray flux delivered to the sample position. Typically, an X-ray mirror is made of fused silica, ZERODUR, CDV SiC, Ni, or Al as substrate and an overlayer of Pt, Au or Rh is coated on the surface[1]. The coating material typically is a high electron density material in order to increase the critical angle of X-ray reflectivity so that the length of the mirror can be reduced. Both the surface contamination, especially the carbon deposition on the mirror, and the surface roughness on the mirror surface,actually, reduce the electron density near the mirror surface which thus reduce also the reflectivity of the X-rays and enhance the unwanted diffusive reflections. Therefore, for a good mirror surface used at X-ray beamline in the synchrotron radiation facility was always polished to within 10 Å of surface roughness and operated under a UHV condition in order to avoid the carbon contamination on the mirror surface, mainly, to eliminate the partial pressure of hydrocarbons. It is well known that the carbon contamination will be a serious problem which limits the effectiveness of the X-ray source especially for the X-ray energies of above the K-edge of carbon. A loss of one or two order of magnitudes of intensity has been reported[2,3]. To avoid the carbon contamination problem, several methods of cleaning the mirror surface using the oxygen glow discharge[4,5] or rf discharge method[6,7,8] had also been developed in order to regenerate the mirror surface either by in-situ or by ex-situ methods. However, for the X-ray energies of above 6 KeV where most of X-ray diffraction/scattering experiments are performed, the problem of carbon contamination is never been emphasized. Part of the reason might be due to the low X-ray absorption in this contaminated layer; the X-rays can easily penetrate through the contaminated layer and be reflected back by the underlying coating heavy materials. Therefore, some of the mirrors for the hard X-ray experiments at synchrotron facilities and research X-ray tubes are operated either at low vacuum (10^{-3} Torr) or even running at atmosphere environment. So far, no strong support was found on the necessity of an X-ray mirror be put under a UHV condition for the usage of hard X-rays. In order to clarify this point, we therefore perform a calculation to elucidate this issue.

II. Method of Calculation:

X-ray mirrors can be used to reflect the X-rays because at incidence angle of less than a critical angle θ_c, the incident X-rays are totally reflected from the surface of the mirror[9,10,11]. The value of critical angle can be derived from the refraction index, n, which can be calculated as follows:

$$n = 1-\delta-i\beta, \tag{1}$$

where δ, β are positive optical constants with values of order of about 10^{-5} or less for the X-ray energies we are interested. These values can be expressed as:

$$\delta = \rho\lambda^2 r_0/2\pi,$$
$$\beta = \lambda\mu/4\pi, \tag{2}$$

where ρ is the effective electron density after subtracting a small fraction of electrons with binding energies greater than the incident X-ray energy. r_0 is the classical electron radius or Thompson scattering length, and μ is the X-ray absorption length. The critical angle for the X-rays reflection can be calculated from the Snell's law which can be approximately written as:

$$\theta_c = (2\delta)^{1/2} \tag{3}$$

In order to reflect X-rays effectively, the X-ray incidence angle must be kept below this critical angle. From Eq. (2) & (3), we can understand that: the coating material on the mirror surface should be a high-Z material such as Pt or Au in order to increase the acceptable angles of incidence X-rays.

We can also see that: for a high energy X-ray, the critical angle will be smaller than that of low energy one. Therefore, the X-ray mirror can be acted as a high energy filter for the white X-rays at fixed incidence angle. In our present study, we choice Pt as our coating material on the mirror surface.

In order to calculate the mirror reflectivity, we have to solve the Maxwell's equation at the mirror surface. One can yield an expression of the Fresnel coefficient, $F_{1,2}$, for the reflection from an ideal smooth surface with semi-infinite bulk material, as a function of incidence angle θ[9]:

$$F_{1,2} = \frac{E_1{}^R}{E_1} = \frac{f_1 - f_2}{f_1 + f_2} \tag{4}$$

where $f_j = (n_j{}^2 - \cos^2\theta)^{1/2}$ with j=1, 2 referred as the first layer (air or vacuum) and semi-infinite mirror material respectively. Then, the Fresnel reflectivity, $I(\theta)/I_0$, can be approximately written as:

$$I(\theta)/I_0 = F_{1,2}F_{1,2}{}^* = \left| \frac{\theta - (\theta^2 - \theta_c - i\beta)^{1/2}}{\theta + (\theta^2 - \theta_c - i\beta)^{1/2}} \right|^2 \tag{5}$$

if the outside layer is in a vacuum environment.

For a real surface, the surface boundary is no longer sharp and smooth, it is convenient to model the in-plane averaged electron density of a simple surface by a Gaussian smeared step from $\rho_1 = 0$ to ρ_2 along the surface normal direction z[10,11]:

$$\rho(z) = \rho_2 \; [1 + \mathrm{erf}(z/2\sigma)]/2, \tag{6}$$

where σ is the root-mean-square average of the surface width, resulting from both the intrinsic width of the interface and mean-square average of the roughness of the surface. The Fresnel reflectivity can then be modified as[11]:

$$I(\theta)/=I_0 = |\, F_{1,2} \exp(-0.5_{q1q2}\sigma^2) \,|^2 \tag{7}$$

where $q_j = 4\pi\sin(\theta_j)/\lambda$, which is related to the normal components of the wave-vectors.

For a multi-layer model[9], the calculated reflectivity is based upon a recursive computation. The Fresnel reflection coefficient of each j layer is defined as $R_{j,j+1} = a_j\ (E_j{}^R/E_j)$ and $a_j = \exp(-i\pi f_j d_j/\lambda)$ where d_j is the layer thickness and E_j is the reflected wave amplitude. The recursion relation:

$$R_{j,j+1} = a_j{}^4 \frac{(R_{j+1,j+2}) + (F_{j,j+1})}{(R_{j+1,j+2}\ F_{j,j+1}) + 1} \tag{8}$$

is then used to compute the reflected intensity by using $I(\theta)/I_0 = R_{1,2}R_{1,2}{}^*$. For the interface between j and j+1 layer, the surface roughness parameter in Eq. (7) is replaced by a random height distribution with a root-mean-square height variation of $\sigma_{j,j+1}$ and Gaussian height-height lateral correlations. Under these conditions, the Fresnel coefficient for the interface between the j and j+1 layer is:

$$F_{j,j+1} = \frac{f_j - f_{j+1}}{f_j + f_{j+1}} C_{j,j+1} \tag{9}$$

where $C_{j,j+1}$ is an approximate correction to the Fresnel coefficient with $C_{j,j+1} = \exp(-0.5 q_j q_{j+1} \sigma_{j,j+1}{}^2)$. It is obvious that the two layer model in Eq(7). is a special case of the multilayer model in Eq(9).

III. Calculation Results:

A. The thickness of the Pt coating:

Assuming that the Pt is coated on the surface of a fused silica with a roughness of 8 Å initially. **Fig.1** shows the calculation of the reflectivity of 4, 8 and 12 KeV X-rays as a function of the thickness of Pt coating at an incidence angle of 6 mrad. The results show that the thickness of Pt layer larger than 100 Å is enough to reflect the properties of a Pt coating mirror.Further deposition of Pt atoms is not necessary.

B. The effect of surface roughness on a clean mirror:

Fig.2 shows the reflectivity of 4,8 and 12 KeV X-rays as a function of surface roughness of a Pt coating mirror at an incidence angle of 6 mrad. The surface are assumed to be clean. The results show that: in order to get 70% of total reflected X-rays at 8 KeV, the surface roughness of the Pt surface must be kept below 33 Å.

C. Incidence angle dependence of a clean mirror:

Fig.3 shows the reflectivity of 4,8 and 12 KeV X-rays as a function of incidence angle. The surface roughness of the Pt coating mirror was assumed to be 8 Å. The results show a strong incidence angle dependence of X-ray reflectivity. For the low energy X-ray, the critical angle is larger than the high energy one, therefore, its reflectivity curve extends to higher incidence angles.

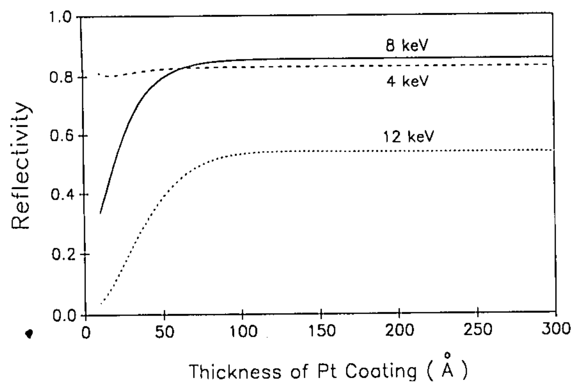

Fig.1 The X-ray reflectivity of a clean mirror as a function of the thickness of Pt coating on fused silica. The roughness of interface between fused silica and Pt layer is assumed to be 8 Å and the incidence angle of X-rays is fixed at 6 mrad. It shows that a Pt coating of 100 Å is sufficient.

D. Energy dependence of a clean mirror:

For a clean Pt coating mirror with a surface roughness of 8 Å, the reflectivity curves as a function of X-ray energy at incidence angle of 4,6 and 8 mrad are shown in **Fig.4**. The result is closed related with the Fig. 3. The X-ray mirror operated at different incidence angles can be used as an energy low pass filter.

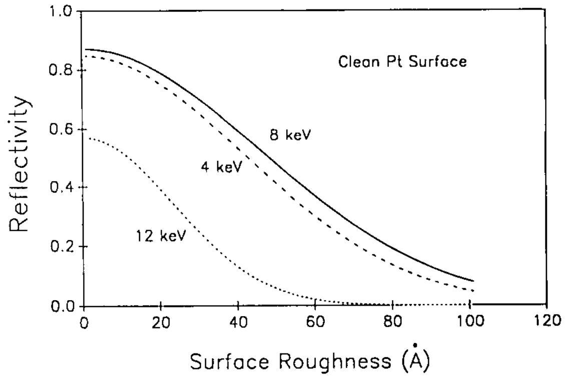

Fig.2 The X-ray reflectivity of a clean Pt coating mirror as a function of the roughness of the mirror surface. The incidence angle is assumed to be 6 mrad.

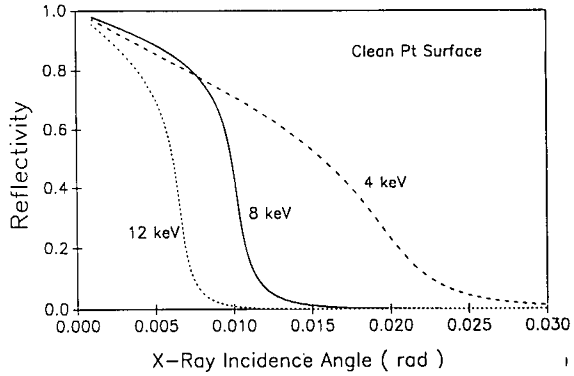

Fig.3 The X-ray reflectivity of a clean Pt coating mirror as a function of X-ray incidence angle. The surface roughness of the Pt coating mirror is assumed to be 8 Å.

E. Effects of carbon contamination:

(i) Thickness dependence:

Fig.5 shows the reflectivity of 8 KeV X-ray as a function of the thickness of carbon layer (the incidence angle is 6 mrad). The roughness between the interface of carbon layer and Pt coating is assumed to be 8 Å. the density of carbon layer can be assumed to be the same as graphite[12]. The surface roughness of the carbon layer is assumed to be 10% of its thickness. From this figure, we can see that: In order to gain 70% of the reflectivity for the 8 KeV X-ray, the thickness of the carbon deposition layer should be kept below 4500 Å. For the 12 KeV X-ray, the slope of reflectivity decrease is smaller than that of 8 KeV X-ray because the absorption coefficient of the carbon layer for 12 KeV X-ray is smaller. For 4 KeV X-ray, the reflectivity seems almost independent of the thickness of the carbon layer. It can be understood by realizing that the 4 KeV X-ray is reflected from the surface of carbon layer at this incidence angle, therefore, it should be independent of the thickness of the carbon layer.

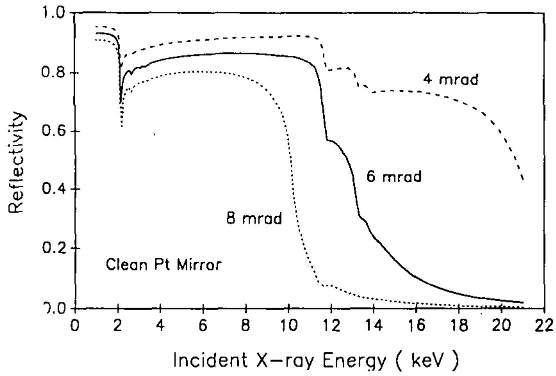

Fig.4 The X-ray reflectivity of a clean Pt coating mirror as a function of X-ray energy. The surface roughness of the Pt coating mirror is assumed to be 8 Å.

It is also worthy to note that the smoothness of the surface of carbon layer will affect the calculation result significantly. For example, **Fig.6** shows the result of this calculation with the surface roughness of the carbon layer of 8 Å, 20 Å and 100 Å. The incident X-ray is kept at 8 KeV and the incidence angle is kept at 6 mrad. We can see that: when the surface roughness of the carbon layer is 8 Å, a clear interference pattern as a function of the carbon thickness can be seen. This interference pattern is then smeared out as the roughness of the surface of the carbon layer is increased. It can be interpreted as follows: When the surface of carbon layer is smooth, the amount of X-ray reflected from the interface between the vacuum and carbon layer is

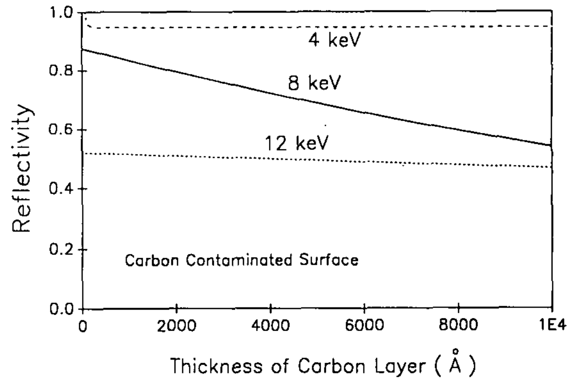

Fig.5 The X-ray reflectivity of a carbon contaminated mirror as a function of the thickness of the carbon layer. The roughness at the interface between carbon layer and Pt coating layer is assumed to be 8 Å and the surface of carbon layer is assumed to be 10% of the thickness of the carbon layer. The incidence angle of X-ray is kept at 6 mrad.

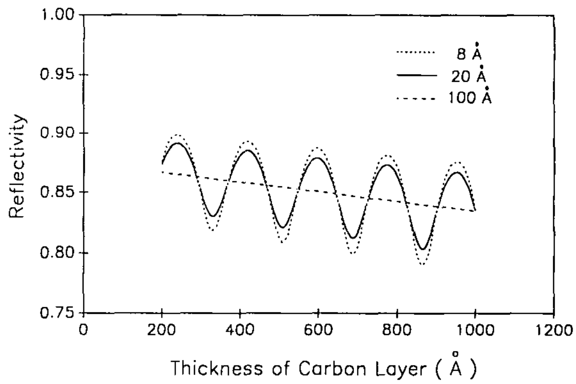

Fig.6 The effect of surface roughness of the carbon layer on the X-ray reflectivity curves. The incidence angle of the X-rays (8 KeV) is 6 mrad.

strong enough to interfere with the X-rays reflected from the underlaying Pt surface, then a clear interference pattern appears. If the surface of the carbon layer is rough enough, most of the X-rays will penetrate through the surface of the carbon layer with reflections mostly taking place at the interface between carbon layer and Pt layer, which makes the X-ray reflection pattern quite similar to that solely from the Pt surface without forming interference pattern.

(ii) Grazing incidence angle dependence:

Fig.7 shows the incidence angle dependence of 8 KeV X-ray reflectivity. The thickness of the carbon layer is 1000 Å. The interface roughness between the Pt and carbon layer is assumed to be 8 Å. In **Fig. 7 a**, the surface roughness is assumed to be 100 Å, and in **Fig.7 b**, 20 Å is assumed. From these calculation results, we can see that at low incidence angle, the X-ray is reflected from the surface of the carbon layer, which produces a high reflectivity than that of the clean Pt coating mirror (see Fig.3) because it avoids a high absorption cross section of the Pt material. For the incdence angle larger than the critical angle of the carbon layer, most of X-rays penetrate into the carbon layer and reflect only through the interface between Pt and carbon layer. A dip at incidence angle of 4.1 mrad for 8 KeV X-ray corresponds to the critical angle of carbon overlayer.

Compared with the Fig.7a and 7b, we can also see that: as the surface roughness of the carbon layer is smaller, an interference pattern is more clearly shown up. The reason is the same as the one in Fig.6.

(iii) Energy dependence:

Fig.8 shows the energy dependence of X-ray reflectivity with a carbon overlayer of 1000 Å and surface roughness of 100 Å. The roughness of the interface between the Pt and carbon layer is assumed to be 8 Å. The grazing incidence angle is fixed at 6 mrad. It is interesting to see that the reflectivity curve consists of a dip located around 5.6 KeV.

This dip can be interpreted as follows: below the 5 KeV, most of X-rays are reflected from the surface of the carbon layer, and above the 6 KeV, the X-rays are mainly reflected from the Pt surface. Around the 5.6 keV, X-rays may only propagate inside the carbon layer, and finally, most of the X-rays are absorbed. This dip is troublesome if an X-ray spectroscopy experiment, such as EXAFS (Extended X-ray Absorption Fine Structure) or anomalous X-ray scattering,is performed around this energy range. It is better to move this dip to another energy region by tilting the mirror to a different incidence angle.

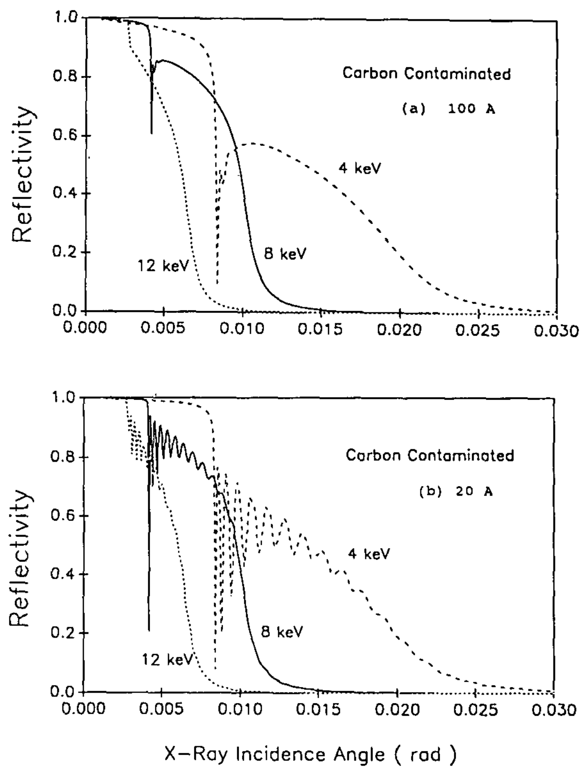

Fig.7 The X-ray reflectivity of a carbon contaminated layer as a function of the X-ray incidence angle. The roughness of the interface between the carbon layer and Pt coating is assumed to be 8 Å. The incident angle is fixed at 6 mrad. (a) The surface roughness of the carbon layer is 100 Å, (b) 20 Å.

Ⅳ. Discussion:

We have calculated the effects of surface roughness and carbon contamination on an X-ray mirror. In order to gain 70% of reflectivity for 8 KeV X-ray, the surface roughness of a Pt coating mirror should be kept below 33 Å, and thickness of the carbon contamination layer should be kept below 4500 Å for the X-ray with an incidence angle of 6 mrad. The current technique of polishing the mirror surface to less than 8 Å is achievable[1]. However, to avoid the carbon contamination needs more efforts. According to the experiment of K. Boller et.al. [3] carried out at synchrotron radiation facility of DORIS (Hamburg, Germany), the deposition rate of the carbon layer is about 100 Å under 100 Amp. Sec of synchrotron white beam irradiation at pressure of above 1×10^{-7} torr. They also found that the deposition rate is proportional to the pressure above the mirror up to the 10^{-7} torr only. It should be pointed out that above the 10^{-7} torr, the coverage of hydrocarbon molecules on the mirror surface will be saturated as one monolayer, thus further high pressure of hydrocarbon gases does not reacted directly on the surface of the carbon deposited mirror. For the mirror of X-ray beamline at the synchrotron in Taiwan, if we assumed the storage ring is operated at an averaged current of 150 mA, and 20 hrs each day, since our flux of low energy X-rays generated from the SRRC wiggler source is approximately 50 times higher than DORIS, to deposit 4500 Å of carbon layer will need only about 50 days (if the mirror surface is maintained at 1×10^{-10} torr and 100°C). Development of the in-situ cleaning techniques of the X-ray mirror in Taiwan will be indispensable. Alternatively, by reducing the incident flux on the mirror surface can also reduce the deposition rate of carbon contamination. An energy filter (typically, several μm of graphite foil) placed before the mirror to absorb most of low energy X-rays will significantly reduce the deposition rate. Placed the mirror after the monochromator to eliminate most of the unwanted white beams can also greatly improve the surface condition of the X-ray mirror. With all the efforts to remove the unwanted X-rays before hitting the mirror surface, the service time of the X-ray mirror can be prolonged for more than 1000 times. It also opens an opportunity of operating the mirror at low vacuum without in-situ cleaning. However, since the mechanism of the carbon deposition is not completely known at this stage, we can not determine the service time of the X-ray mirror accurately right now. However, the calculation in this work does show that it will affect the configuration of an X-ray beamline design deeply.

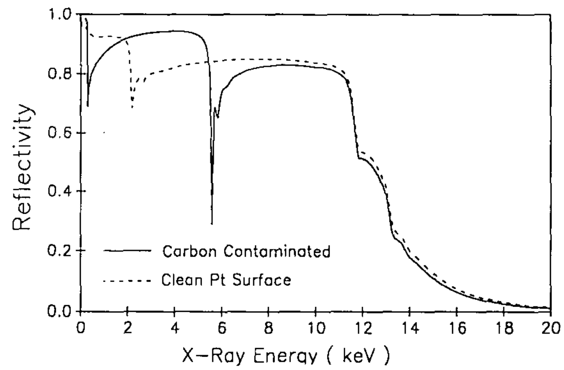

Fig.8 The X-ray reflectivity of a carbon contaminated mirror as a function of the incident X-ray energy. The thickness of the carbon layer is assumed to be 1000 Å and the X-ray incidence angle is kept at 6 mrad.

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通　知

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