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ACCURATE DETERMINATION OF FILM THICKNESS BY LOW-ANGLE X-RAY REFLECTION*

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A modified Bragg law for straightforward determining the film thickness by low-angle X-ray reflection was derived based on the geometrical optical theory. We showed that this modified Bragg law and its inference formulae could be used to accurately determine the thickness of the monolayer or multilayer film. Furthermore, the modified Bragg law for determining the superlattice period presented earlier by others can be derived from the above modified Bragg law. The similar inference formulae were also given. The precision in determining the film thickness and/or superlattice period by the above formulae was discussed.

Keywords: film, modified Bragg law, X-ray reflection
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I. INTRODUCTION

In film physics, the film thickness is often desirable, both in applied and basic researches, to be determined rapidly and accurately. So far, the most widely-used methods to determine the film thickness have been the X-ray reflection or diffraction technique[1−3] because they are nondestructive and reasonably accurate. For example, the positions of interference fringes observed in a reflectivity curve are used to determine the film thickness by the modified Bragg equation[4]

$$\sin^2\theta_i = \theta_c^2 + (n_i + \Delta n)^2\lambda^2/4t^2, \qquad (1)$$

where θ_i is the position of the maximum or minimum of the observed ith interference fringe, θ_c the critical angle for total reflection, n_i an integer, $\Delta n = 1/2$ and 0 are for the maximum and minimum respectively, and λ is the X-ray wavelength. The film thickness can be determined from the slope of $\sin^2\theta_i$ versus $(n_i+\Delta n)^2$ curve. The superlattice period can be accurately determined by a modified Bragg law suggested by Sugawara *et al.*[5]

$$d = \Lambda\lfloor 1 - (1 - n_{\rm SL})/\sin^2\theta_n \rfloor, \qquad (2)$$

where $d = n\lambda/2\sin\theta_n$, Λ is the superlattice period, and $n_{\rm SL}$ the average X-ray refraction index of the superlattice. However, the determination of the film thickness (or superlattice period) by Eq.(1) (or Eq.(2)) usually needs all the interference peaks (or diffraction peaks). Moreover, it is rather difficult to determine the thickness of thicker films by Eq.(1), because of the difficulty in judging the interference order close to the critical angle.

In recent years, the computer simulation of X-ray reflectivity or high-angle X-ray diffraction was widely applied to thin films.[5−7] The accurate and detailed structural information can be obtained by modeling the film structures and comparing the calculated X-ray intensity of the modeled structures with experimental data. Usually, it is necessary to consider many parameters and spend much time to do the simulation work.

Considering that the kinematical approach has the advantage of computation speed over the more rigorous dynamical theory, in this paper we derived a modified Bragg law and its various forms in conjunction with the geometrical optical theory. One can conveniently determine the film thickness by the modified law and its inference formulae. The precision can be guaranteed in comparison with the reflectivity calculation or other methods.

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II. OPTICAL THEORY OF FILM

According to the geometrical optical theory, the optical path difference l between the X-ray beams reflected at the film surface and the substrate is given by

$$l = 2nD\cos\alpha \tag{3}$$

where n is the refractivity, D the film thickness and α the refraction angle. For multilayers, l is represented by

$$l = 2\sum_i n_i d_i \cos\alpha_i = 2nD\cos\alpha, \tag{4}$$

where, n_i, d_i and α_i are the refractivity, film thickness, and refraction angle of each sublayer respectively; $n\cos\alpha = \sum_i \frac{d_i}{D} n_i \cos\alpha_i$, in which n is the mean refractivity of the multilayer, and D the total thickness.

With Snell law, $n_0 \sin\alpha_0 = n\sin\alpha (n_0 = 1$ for air), we have

$$l = 2D\sqrt{n^2 - \cos^2\theta}, \tag{5}$$

where θ is the X-ray incidence angle. As is well known, the interference occurs in the case of $l = m\lambda$ (m=1,2,3...). Then we have

$$L = D\sqrt{\frac{(1-n^2)}{\sin^2\theta_m}}, \tag{6}$$

where $L = \dfrac{m\lambda}{2\sin\theta_m}$. In general, the dispersion part of the complex refractive index is $\delta \sim 10^{-5}$ for hard X-ray, one can reasonably consider $1 - n^2 \approx 2\delta$. Then, it can be seen that Eq.(6) is in accordance with the equation used by Kiessig[8]. Plotting L^2 versus $\sin^{-2}\theta_m$, one can conveniently determine not only the film thickness by extrapolating the line to the L^2 axis, but also the mean refractivity from the slope.

In the case of $1 - n^2 \ll \sin^2\theta_m$, by expanding the square root, Eq.(6) can be further written as

$$L = D[1 - (1-n^2)/2\sin^2\theta_m]. \tag{7}$$

According to Eq.(7), the film thickness can be determined directly by extrapolating the $L \sim \sin^{-2}\theta_m$ curve to the L axis, which has the advantage of speed over Eq.(1) which determines the film thickness from the slope of $\sin^2\theta_i \sim (n_i + \Delta n)^2$ curve. From Eq.(7), one can clearly see that the Bragg law, $D = \dfrac{m\lambda}{2\sin\theta_m}$, is valid only in the case of $\dfrac{1-n^2}{2\sin^2\theta_m} \ll 1$. Furthermore, from Eq.(7), one can obtain various forms for determining the film thickness, e.g.

(1) $$D = \frac{(k-m)\lambda}{2(\sin\theta_k - \sin\theta_m)\left(1 + \dfrac{1-n^2}{2\sin\theta_k \sin\theta_m}\right)}, \tag{8}$$

where k and θ_k are the interference order and angle respectively. Theoretically, when $1 - n^2 \leq \sin\theta_k \sin\theta_m/50$, the film thickness can be determined by

$$D = \frac{(k-m)\lambda}{2(\sin\theta_k - \sin\theta_m)} \tag{9}$$

with smaller error. This formula is very helpful for determining the thickness of thicker films because it is usually rather difficult to accurately judge the interference orders.

(2) Since the film thickness (D_{mk}) determined by Eq.(9) is larger than the real thickness (D_{real}) by $\dfrac{1-n^2}{2\sin\theta_k\sin\theta_m}$, while the film thickness D_m (or D_k) directly determined by $\dfrac{m\lambda}{2\sin\theta_m}$ (or $\dfrac{k\lambda}{2\sin\theta_k}$) is smaller than the real thickness by $\dfrac{1-n^2}{2\sin^2\theta_m}$ (or $\dfrac{1-n^2}{2\sin^2\theta_k}$). Thus, the average thickness calculated from the values given by Eq.(9), $\dfrac{m\lambda}{2\sin\theta_m}$, and $\dfrac{k\lambda}{2\sin\theta_k}$ will have a much smaller error

$$D_{\text{average}} = \frac{D_m + D_k + 2D_{mk}}{4}. \tag{10}$$

(3) If the interference order and angle are known, the film thickness can also be determined by

$$D = \frac{\lambda\sqrt{k^2 - m^2}}{2\sqrt{\sin^2\theta_k - \sin^2\theta_m}}. \tag{11}$$

For superlattices, the total film thickness $D = N\Lambda$, where N is the number of the superlattice period and Λ the superlattice period. It is reasonably to consider that the first Bragg peak corresponds to the Nth Kiessig interference peak. Then, from Eq.(7), we obtain

$$L_S = \Lambda[1 - (1-n^2)/2\sin^2\theta_M], \tag{12}$$

where $L_S = M\lambda/2\sin\theta_M, \theta_M$ is the Bragg peak position due to the superlattice structure, M the order of Bragg peak. Usually $1 - n^2 \approx 2\delta = 2(1-n)$ for hard X-rays, one can see that Eq.(12) is in good agreement with the modified Bragg law presented by Sugawara *et al.*[5] Similar to Eq.(9), when $1 - n^2 \leq \sin\theta_K \sin\theta_M/50$, the superlattice period can also be directly determined by

$$\Lambda_{KM} = \frac{(K-M)\lambda}{2(\sin\theta_K - \sin\theta_M)} \tag{13}$$

with smaller error, where K and θ_M are the Bragg order and the angle of low-angle X-ray diffraction respectively. Certainly, the superlattice period can also be determined by

$$\Lambda = \frac{\lambda\sqrt{K^2 - M^2}}{2\sqrt{\sin^2\theta_K - \sin^2\theta_M}} \tag{14}$$

or given by an average of the values from Eq.(12), $\Lambda_M \left(= \frac{M\lambda}{2\sin\theta_M}\right)$ and $\Lambda_K \left(= \frac{K\lambda}{2\sin\theta_K}\right)$.

III. EXPERIMENTAL

Tantalum monolayer film and $[Co/Pd]_{20}$ superlattices were deposited on water-cooled single-crystal silicon substrates by magnetron sputtering separately. All of the samples were prepared with a working pressure of 0.55 Pa and a background pressure of 3.7×10^{-5} Pa. The reflectivity measurements were performed on a diffractometer attached to a 12 kW rotating anode X-ray generator.

IV. COMPARATIVE DISCUSSION

Curve A in Fig. 1 is the reflectivity curve of a Ta monolayer film. One can see several oscillation peaks due to the interference between the X-ray beams reflected at the film surface and the substrate. The film thickness was determined to be 13.08nm, 12.96nm, and 12.98 nm by using the $L \sim \sin^{-2}\theta_m$ curve (Fig.2), Eq.(10) and Eq.(11) respectively. By simulating the reflectivity curve (the dashed curve in Fig.1), we determined the film thickness to be 13.2 nm. Comparing the value from the simulation with the above results, we can see that the difference is within 2%. This indicates that the film thickness can be determined accurately by $L \sim \sin^{-2}\theta_m$ curve or Eq.(10) or Eq.(11). It is noted that the film thickness determined from Eq.(1) is 13.61nm, obviously larger than any value above. This may be caused by the approximation in deriving Eq.(1). It should be pointed out that when using Eq.(10) or Eq.(11) to determine the film thickness, it is better to choose the interference peaks far from the critical angle. This is because the interference peaks close to the critical angle may cause larger error. For instance, the film thickness determined from Eq.(11) using the second and third Bragg peaks is 12.27nm, about 7% smaller than the simulation value.

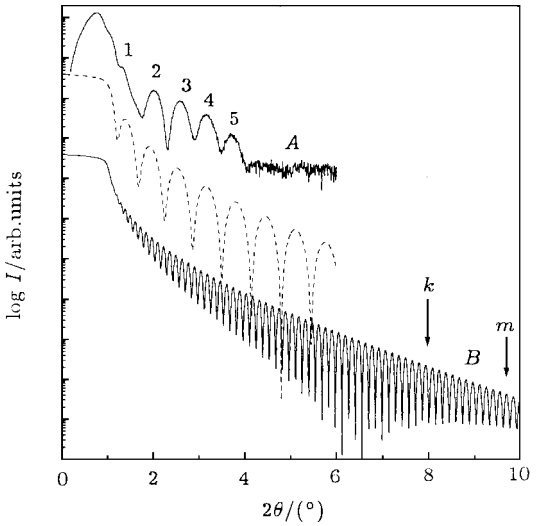


Fig.1. The reflectivity curves of Ta monolayer, where the dashed curve is the calculated result of curve A Curve B is a calculated reflectivity curve of 60 nm Ta film.

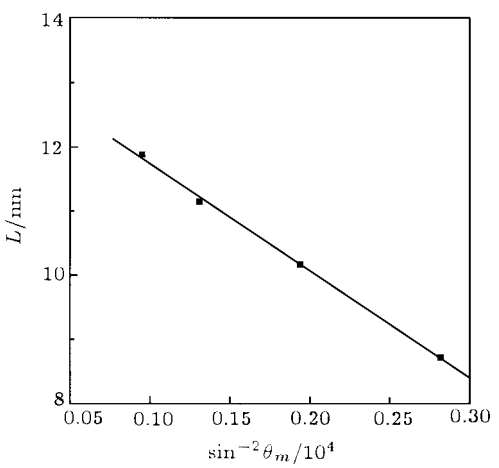


Fig.2. The curve of L versus $\sin^{-2}\theta_m$, in which the solid curve is the linear fitted result.

For the thicker film, in general, it is rather difficult to judge the interference order because the interference peaks close to critical angle cannot be clearly seen. As an example, curve B in Fig.1 shows the calculated reflectivity curve of Ta monolayer film with a thickness of 60 nm (there will impossibly be so obvious interference peaks close to critical angle in actual reflectivity curve). k and m are determined to be 51 and 63 directly from the curve respectively. However, it is not the case. According to Eq.(9), the film thickness was determined to be 60.81 nm from the kth and mth interference peaks. Taking this value, we obtained m=66.7 by Bragg law $(\frac{m\lambda}{2\sin\theta_m})$. Since the thickness determined directly by Bragg law should be a little smaller than that determined by Eq.(9) in small angle region, it is reasonable to infer that m should be 66,

then k should be 54. After knowing the interference orders, the thickness was further determined to be 60.41nm, 60.46 nm, and 60.38 nm by the $L \sim \sin^{-2}\theta_m$ curve, Eq.(10) and Eq.(11) respectively, all of which are different from the real thickness within 1%, indicating that the film thickness can be determined more accurately by the present method. Furthermore, the film thickness was determined to be 60.75 nm by Eq.(1). In comparison with the real film thickness (60 nm), it can be seen that the value determined by Eq.(1) has a larger error than those determined by the $L \sim \sin^{-2}\theta_m$ curve, Eq.(10) and Eq.(11).

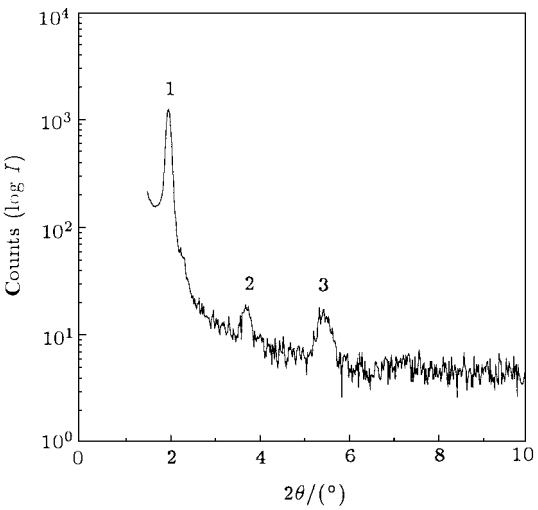


Fig.3. The reflectivity curve of $[Co/Pd]_{20}$ superlattices.

Figure 3 is the low-angle X-ray diffraction results of magnetron-sputtered $[Co/Pd]_{20}$ superlattices. Three Bragg peaks corresponding to $2\theta = 2.03°$, $3.72°$ and $5.46°$ are seen in the curve. Using Eq.(2), the superlattice period (Λ) is accurately determined to be 4.92 nm by extrapolating the $L_S \sim \sin^{-2}\theta_M$ curve to the L_S axis (not given here). On the other hand, the Bragg law directly gives the values of the period, $\Lambda_2 = 4.74$ nm and $\Lambda_3 = 4.85$ nm and Λ_{23}=5.08 nm from the second and third Bragg peaks, respectively. As expected, the value directly determined by the Bragg law is smaller than the above value 4.92 nm (within errors 3.7% and 1.4% respectively), while the value determined by Eq.(13) is higher (within error 3.4%). However, the superlattice period is determined to be 4.94 nm from the average of Λ_2, Λ_3 and Λ_{23}, only 0.4% different from 4.92 nm. The superlattice period determined by Eq.(14) is 4.93 nm from the first and third Bragg peaks. The difference among the above values is within 1%, indicating that the superlattice period is also determined accurately by Eq.(14) or the average of Λ_2, Λ_3 and Λ_{23}.

It should be pointed out that the most effective method for accurate determination of the film thickness or superlattice period is the extrapolation of the straight line $L \sim \sin^2\theta_m$ or $L_S \sim \sin^2\theta_M$ because such a procedure which uses all the experimental data makes error the smallest. But in the case that at least two well-defined peaks far from the critical angle are observed, it is also effective to determine the film thickness or superlattice period by the inference formulae derived from the modified Bragg law.

V. CONCLUSION

We have derived a modified Bragg law and its various forms based on the geometrical optical theory. One can accurately determine the film thickness and/or superlattice period by this modified Bragg law and its corresponding inference formulae. The accuracy of the result is extremely satisfactory compared with the reflectivity calculation or other methods.

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