Study on quasiperiodic Ta/Al multilayer films by x-ray diffraction

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(Received 28 May 1991; accepted for publication 14 August 1991)

Quasiperiodic (Fibonacci) Ta/Al multilayer films with Ta(110) and Al(111) textures were fabricated by magnetron sputtering. The structure of the multilayers was characterized in detail by x-ray diffraction. The diffraction peaks at low and high angles can be indexed by the projection method from the high-dimension periodic structure. The experimental results were in good agreement with the numerical calculation using the model for the compositionally modulated multilayers. The diffraction spectrum of the quasiperiodic Ta/Al multilayers is totally different from that of periodic structure, and the possible application of Fibonacci films as optical elements in a soft x-ray region is discussed.

In the past two decades, artificial multilayers have been fabricated and investigated, which opens an area to the development of new materials that do not exist in nature.[1–3] These new materials have been shown to exhibit unusual behavior of many physical properties, depending on the layer thickness of the superlattice. The main reason for the versatility of multilayers is that layer thickness can be intentionally changed at length scales ranging from a few angstroms to a few hundered angstroms. In the last few years, the interest in multilayers was rekindled in the fields of soft x-ray and UV optics.[4,5] With the increasing availability of synchrotron radiation sources, suitable high-Z/low-Z multilayers can be used as optical elements for soft x-ray and vacuum ultraviolet. The wellknown examples are W/C,[6] Nb/Al,[7] and Ta/Al[8] combinations which have been well studied. However, the investigations have been restricted to periodic structures.

The Fibonacci sequence can be obtained by repeated operations of the concurrent substitution rules $A \rightarrow AB$ and $B \rightarrow A$, in which the ratio of two incommensurate intervals (Da and Db) is equal to the golden mean $\tau = (\sqrt{5} + 1)/2$. According to the projection method, the Fourier spectrum of Fibonacci lattices consists of δ function peaks at

$$k(n,m) = 2\pi D^{-1}(n + m\tau) \tag{1}$$

where $D = \tau Da + Db$ is the average lattice parameter and n, m are integers. Even for $Da/Db \neq \tau$, the quasiperiodic properties are preserved due to the topological equivalence between a square and a rectangle in projection method. In 1985, Merlin *et al.*[9] reported the first realization of a quasiperiodic GaAs-AlAs superlattice. Since then many experiments on quasiperiodic superlattices have been reported.[10–12] However, only a few studies on x-ray diffraction of these structures in the low angle region has been reported[10] and no attention has been paid to the high-Z/low-Z quasiperiodic multilayers.

In this letter, the fabrication and structural characterization of quasiperiodic Fibonacci Ta/Al multilayers are reported. The possible application for soft x-ray optics is also discussed.

The quasiperiodic Ta/Al multilayer films were grown by magnetron sputtering. The procedure involves defining two distinct building blocks and ordering them in a Fibonacci sequence. In a typical sample which we prepared, the building blocks A and B consist nominally of (12.6 Å Ta)–(34 Å Al) and (12.6 Å Ta)–(17 Å Al), so the average lattice parameter $D = \tau Da + Db \cong 105$ Å. The samples we studied consist of thirteen generations of Fibonacci sequence and they are about 1.5 μm in thickness.

The multilayer films were characterized by x-ray diffractometry. A 12 kW Rigaku rotating anode x-ray source was used. The measurements were made both near the Bragg peaks of Ta and Al at $2\theta = 38.5°$ and at the grazing angles of incidence ($0.5° \leqslant 2\theta \leqslant 10°$). Both types of measurements are significant for the characterization of these samples. The scattering vector was kept normal to the surface for this diffraction pattern. In the high angle region, the main diffraction peak was found [shown in Fig. 1(a)] representing the reflections from the bcc Ta(110) and fcc Al(111) planes which have an equal interlayer spacing of $a = 0.2338$ nm, and no other main peaks were found. From this, the sample is dominated by crystalline Ta and Al with texture Ta(110) and Al(111). On both sides of main Bragg reflection, there are some Fibonacci satellite peaks, which can be indexed as $\left[{n \atop m} \right]$ [shown in Fig. 1(a)].

In the low angle region, at least fifteen harmonics have been observed [shown in Fig. 2(a)]. These Fibonacci peaks can also be indexed and labeled by $\left[{n \atop m} \right]$. Their relative intensities, peaks positions, indexes, and scattering vectors are listed in Table I. All experimental values of k are in good agreement with calculated ones from Eq. (1), and

$$k(a_{n+1}, a_{n+2}) = k(a_n, a_{n+1}) + k(a_{n-1}, a_n) \tag{2}$$

is satisfied where a_n is Fibonacci number defined by $a_n = a_{n-1} + a_{n-2}$ with $a_0 = 0$ and $a_1 = 1$. It reflects the self-similarity of the reciprocal lattice. On the other hand, it should be noted that in Table I there are a few peaks with high relative intensities. Compared with the experimental results for the periodic Ta/Al structure,[8] the intensity of this strongest peak (SP) is almost equivalent to that of first order of periodic Ta/Al multilayers. The relative intensities of the second strongest peak (SSP) labeled by $\left[{1 \atop 2} \right]$ and the third strongest peak (TSP) labeled by $\left[{0 \atop 1} \right]$ are much higher than 2 order and 3 order of periodic structures. In periodic Ta/Al multilayers, the intensities of 2 order and 3

0003-6951/91/452512-03$02.00

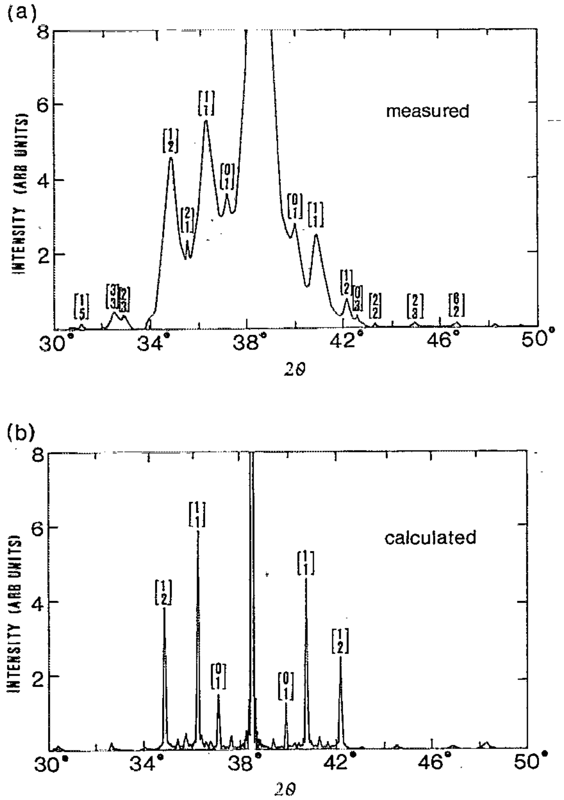

FIG. 1. The θ–2θ scan of x-ray diffraction in the high angle region for quasiperiodic Ta/Al with $D = 105$ Å, Cu K_α radiation. (a) Measured, (b) calculated.

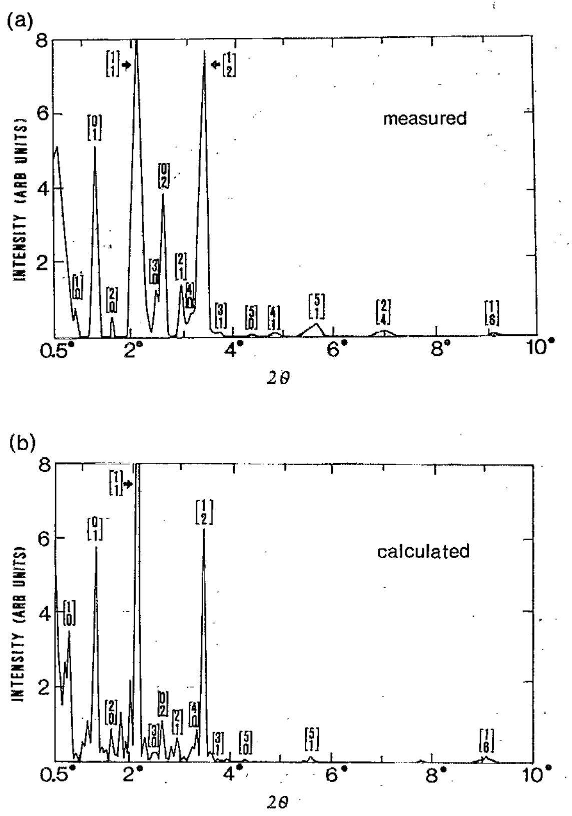

FIG. 2. The θ–2θ scan of x-ray diffraction in the low angle region for quasiperiodic Ta/Al with $D = 105$ Å, Cu K_α radiation. (a) Measured, (b) calculated.

order peaks are lower than 10% of the strongest (1 order) one. However, in the quasiperiodic multilayer the relative intensity of SSP is more than 40% of SP and that of TSP is more than 20% of the SP. Particularly, SSP is far away from $\theta = 0$ [shown in Fig. 2(a)]. The quasiperiodic order gives rise to all of these phenomena which may be usable for some special cases in soft x-ray optics.

In order to obtain more information, the x-ray diffraction patterns were numerically simulated. The model for the compositionally modulated multilayer was used for the simulation. The calculation method was the same as those we used for periodic Ta/Al multilayer.[8] But the sequence is different from periodic multilayer. The results of the fitting calculation are shown in Figs. 1(b) and 2(b) in which the degree of fluctuation in this sample is less than 2% and the coherence length perpendicular to the film is about 100 nm. The experimental data are consistent with the numerical calculation for both scattering intensities and peak positions.

In summary, in the quasiperiodic Ta/Al multilayer,

TABLE I. Diffraction and calculation data in the low angle region for Ta/Al multilayer with $D = 105$ Å, Cu K_α radiation.

Index (n,m)	Peak position 2θ(degree)	Observed intensity (arbitrary units)	$k = 4\pi\lambda_x^{-1}\sin\theta$ (Å^{-1})	$k = 2\pi D^{-1}(n + m\tau)$ (Å^{-1})
10	0.88	31 152	6.259×10^{-2}	5.984×10^{-2}
01	1.30	205 548	9.246×10^{-2}	9.682×10^{-2}
20	1.62	22 089	0.1152	0.1197
11	2.12	333 493	0.1508	0.1567
30	2.48	50 400	0.1764	0.1795
02	2.64	152 881	0.1878	0.1936
21	2.96	57 270	0.2105	0.2165
40	3.30	152 343	0.2347	0.2394
12	3.46	321 958	0.2461	0.2535
31	3.74	4952	0.2660	0.2763
50	4.28	877	0.3043	0.2992
41	4.68	2376	0.3328	0.3362
51	5.64	13 968	0.4010	0.3960
24	7.00	4812	0.4976	0.5070
16	9.06	695	0.6437	0.6408

the high contrast of the refraction index between Ta and Al is well satisfied. The experiments showed that this structure has the high reflectivities at wavelength $\lambda_x = 1.54$ Å. It is well known that one of the most important applications of high-Z/low-Z metallic multilayers is to use them as reflectors for soft x rays and UV radiation. The high reflectivities of multilayers in the soft x rays and UV region at near normal angles of incidence is the primary basis as optical elements in advanced instrumentation. So far, considerable progress has been achieved in using periodic multilayers as reflectors at photon energies of 50 to 583 eV. Some additional improvements are possible with quasiperiodic designs. From the diffraction pattern of quasiperiodic Ta/Al multilayers, there are more than one strong peak in the low angle region at the wavelength $\lambda_x = 1.54$ Å and the possible performance at longer wavelength can be precited. Particularly, the strong peak labeled by $[\frac{1}{2}]$ has large incidence angle. Therefore, normal incidence angles can be chosen in the case of high photon energies in soft x-ray region. For instance, a quasiperiodic multilayer with the average lattice parameter $D = 32.83$ Å can be fabricated and used as the reflector of photon energy of 800 eV. This multilayer, in which the second strongest peak of diffraction is considered to be indexed by $[\frac{1}{2}]$, will have the high reflectivity in $\lambda_x = 15.5$ Å at near normal incidence angle $(\theta = \arcsin)(\lambda_x(n + m\tau)/2D) \cong 90°$, where $n = 1$ and $m = 2$). In contrast, if a periodic multilayer is used on the same purpose, its modulation wavelength will be 7.75 Å or so and obviously its fabrication will be difficult. On the other hand, much better reflector performance in a multilayer consisting of two materials can be obtained by reducing the relative thickness of the more highly absorbing material. It causes the overall absorption per unit length to be reduced so that a greater number of interfaces can contribute the reflection process. In the region of high photon energies, the small modulation wavelength in periodic multilayers is required. Thus, it seems to be difficult to reduce the relative thickness of the high-Z element. But, it may be possible by designing of quasiperiodic high-Z/low-Z multilayers.

This work is supported by National Science Foundation of China.

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