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Structural study of multilayered vanadium/nickel superlattices

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Abstract

We have studied the microstructure of V/Ni metallic superlattice, using x-ray and neutron diffraction. We find a sharp and broad rocking curves around the first order Bragg peak, and attribute them to a columnar structure which gives rise to two modulation structures; one the ordinary layered structure within the columns and the other the averaged modulation structure which produces the sharp rocking peak.

Introduction

Metallic multilayered systems¹ have recently attracted much interest from physicists and engineers because of the discovery of novel magnetic² and superconducting properties,³ and the possibility to apply them as x-ray^{4,5} and neutron⁶ mirrors, polarizers and monochromators.

The study of the structural properties⁷ of these modulated superlattices is fundamental for the understanding of other properties. The molecular beam epitaxy technique⁸ is now commonly used to prepare semiconductor multilayers, whereas sputtering⁹ is often applied to prepare metallic ones. In this paper we present the study of the structural properties of V/Ni system, prepared by sputtering using x-ray and neutron diffraction. Columnar structure has been found with growth direction Ni(111) and V(110) tilted by $\sim 1^\circ$ from the normal to the substrate surface. The structure is characterized by a sharp and a broad scattering profile in the azimuthal direction (rocking curve) near the first order Bragg peak of the modulation structure.

Sample Preparation

V/Ni multilayer samples were prepared by sequential sputtering deposition of V and Ni using high rate magnetron sputtering on room temperature, single crystal (0 and 90°) sapphires and glass slide. The sputtering rates for V and Ni were kept around 10 Å/sec. The base vacuum before the introduction of the Ar sputtering gas (~ 10 mtorr) was $2-5 \times 10^{-7}$ torr. The distance between the substrate and sputtering gun is kept close to 4 inches, and the substrates are alternately moved from one beam to the next thus depositing alternating layers. Equal thickness bilayer structure was maintained throughout this study (i.e., $d_{Ni}=d_V=A/2$, where d_{Ni} and d_V are the layer thickness of Ni and V in one period). Three different sets of samples with varying number of bilayer N were prepared ($A = 30, 50$, and 100 Å).

Experimental Results

The x-ray experiments were made using a Rigaku D/Max system with 2-kW Cu x-ray source (~ 1 mm diameter x 10 mm height) and a pyrolytic graphite (PG) crystal analyzer. Neutron diffraction was performed using unpolarized neutrons (1" diameter x 1" height) from liq. H₂ cold source at Brookhaven National Laboratory with two PG monochromators. All diffraction experiments were carried out at room temperature.

0/20 scans at large and small scattering angle regions were used to characterize the modulated structure. Fig. 1 shows the high angle Bragg peaks modulated by the superlattice structure, for $A = 30, 50$ and 100 Å. The growth consists of stacks of Ni(111) and V(110) atomic planes in the perpendicular direction of the film plane. The existence of these superlattice peaks shows that the modulation structure has long range order. An example of the small angle neutron scattering 0/20 scans for $A = 100$ Å, $N = 50$

Fig. 1 High angle 0-20 x-ray diffraction intensity spectra for equal layer thickness V/Ni superlattices with N number of bilayers (a) N=125, (b) N=100, and (c) N=50.

Fig. 2 Small-angle 0-20 neutron diffraction intensity spectrum for a V/Ni superlattice, with N=50 and $\Lambda=100\text{\AA}$.

sample is shown in Fig. 2. Bragg peaks up to the third order are observed indicating that the layered structure is well defined.

Reflection ω -scan "rocking curve" (i.e. for fixed 2θ value, the sample is rotated with respect to the incident beam) are used to study further the microscopic structure of the system. Fig. 3 shows a typical rocking curves for the $\Lambda = 30\text{\AA}$, $N = 200$ sample at low (around the first Bragg peak $2\theta \sim 2.74^\circ$) and high (around the central peak $2\theta \sim 43.2^\circ$) angles. In Fig. 3(a), sharp ($\Delta\omega \approx 0.05^\circ$) and broad peaks ($\Delta\omega \sim 2^\circ$) are superimposed. High angle rocking curve in Fig. 3(b) gives the mosaic spread angle, $\Delta\omega \sim 10^\circ$.

To characterize the sharp and broad peaks we made two additional sets of experiments. First, we mapped out the x-ray intensity of the first order Bragg peak in k -space. A series of rocking curves were taken with small increments ($\sim 0.02^\circ$), of 2θ around $2\theta \sim 2.74^\circ$. The sharp peak is centered at 2θ , and has a full width at half maximum (FWHM) $\Delta 2\theta \sim 0.12^\circ$ and $\Delta\omega \sim 0.05^\circ$ in the radial and azimuthal direction respectively. On the other hand, the broad peak position is at $2\theta \sim 2.66^\circ$, with $\Delta 2\theta \sim 0.12^\circ$ and $\Delta\omega \sim 2^\circ$, which looks like a disc in k -space. Second, we carried out the same experiment as described in (1) in four different geometries to characterize the structural anisotropy

Fig. 3 (a) Small-angle (around $2\theta_0 = 2.74^\circ$) and (b) the high-angle (around $2\theta_0 = 43.20^\circ$), rocking curves for a V/Ni superlattice with $\Lambda = 30 \text{ \AA}$ and $N=200$.

parallel to the layers (x-y). The sharp peak was independent of the orientation of the sample whereas the broad peak changed in a systematic way with sample orientation.

Fig. 4 Transmission 0-2 θ x-ray diffraction intensity spectrum for V/Ni superlattice with $\Lambda = 30 \text{ \AA}$ and $N = 200$.

In-plane domain size (or the coherence length) was estimated from transmission $\omega/2\theta$ scan near $V(110)$ peak (see Fig. 4). $\Delta 2\theta$ at FWHM $\sim 2^\circ$ is comparable to $\Delta\epsilon$ of the broad peak at small angle, and leads to a grain size of $d \sim 50$ Å.

Fig. 5 (a) X-ray peak intensity of the sharp rocking curve, and (b) x-ray integrated intensity of the broad rocking curve versus the number of bilayers, N for $\Lambda = 30$ Å γ /Ni superlattice.

Furthermore the x-ray intensity around the first order Bragg peaks for both sharp and broad ω -scan were studied as a function of N , for fixed Λ . Typical results are shown in Fig. 5 for $\Lambda = 30$ Å. Fig. 5 (a) shows that the sharp peak intensity increases up to $N \sim 45$ and then decreases rapidly. The sharp peak width remains constant as a function of N ($\Delta\omega_{FWHM} = 0.05^\circ$). The broad peak integrated intensity $I_{int} (\sim I_0 \times \Delta 2\theta)$ versus N is shown in Fig. 5(b). I_{int} increases linearly with N and saturates around $N \sim 50$.

We will show later that all these results can be explained self-consistently using a columnar model for the growth.

Around the first order Bragg diffraction peak finite size oscillations are observed in the $\omega/2\theta$ scans as shown in Fig. 6. This oscillatory behavior have been observed up to $N \sim 20$, indicating that the sharp peak arises from a region of 600 Å ($\approx 20 \times 30$ Å) close to the substrate. As the thickness increases, the rest of the layers do not contribute to the sharp peak. This mechanism will explain the results of Fig. 5(a) and (b).

Fig. 6 Small angle 0-2θ x-ray diffraction intensity spectrum for V/Ni superlattice with $\Lambda = 30 \text{ \AA}$, $N = 20$.

Discussion

From the high angle scattering data (Fig. 1), it is possible to derive the modulation wavelength Λ , interatomic spacing $d_{\text{Ni}(111)}$, $d_{\text{V}(110)}$, the number of atomic planes in the unit cell N_{Ni} , N_{V} and interface information (i.e., the degree of roughness at interfaces, by least-square fitting the data to models using the kinematical model.^{10,11}

Here we are mainly concerned with the microstructures of the V/Ni system. Fig. 3 shows the two distinct rocking curve lineshapes, a sharp ($\Delta\omega \sim 0.05^\circ$), and a broad ($\Delta\omega \sim 2^\circ$) one. To our knowledge this is the first observation of this kind of behavior. A model which can explain the data in a consistent fashion is as follows: the multilayered film grows in a columnar-like fashion with textured growth almost perpendicular to the substrate surface, and random orientation in the xy plane of the substrate. Fig. 7 shows growth with the column axis slightly tilted (about 1°) from the normal direction the film.

This tilt has been substantiated with a series of experiments performed by rotating the sample around the z-axis. Although the tilt origin is not clear at present, we believe its related to a slight tilt in our substrate holder platform. The intensity distribution of the broad peak in k-space is disc-like (Fig. 8 (b)) with $\Delta 2\theta^B = 0.12^\circ$, $\Delta\omega^B = 2^\circ$ (i.e., $\Delta k_v = 0.0085 \text{ \AA}^{-1}$ and $\Delta k_{h12} = 0.14 \text{ \AA}^{-1}$) which leads to a columnar structure (Fig. 8(a)) with $L \sim 740 \text{ \AA}$ and $d \sim 50 \text{ \AA}$.¹² The sharp peak on the other hand is obtained from the average modulation structures with periodicity Λ' , where the interfaces have terrace-like roughness as shown in Fig. 7. This new periodicity Λ' ($\Lambda' < \Lambda$) will be well defined as long as the column axes are parallel to each other.

This model is capable of explaining all experimental results: 1) the oscillatory behavior (Fig. 6) is due to finite size effects of the modulation structure Λ' with a coherence length $\sim 700 \text{ \AA}$, 2) The rapid decrease of the sharp peak intensity with N (Fig. 5 (a)) is due to the increasing spread in the column growth direction beyond $\sim 1000 \text{ \AA}$. Further growth above $\sim 1000 \text{ \AA}$ acts as an absorber for the sharp peak which therefore decrease in intensity. (3) The saturation in the broad peak intensity vs N (Fig. 5b) is due to the finite coherence length which is smaller than the thickness of the film.

Conclusion

We have studied the microstructure of V/Ni multilayered system using x-ray and neutron diffraction. The experimental evidence implies that the film growth is columnar in this multilayered system. The structure gives rise to two distinct modulation periodicities; one crystalline where the periodicity is along the column axes, the other due to the averaged modulation of the interfaces which is well-defined as long as the column growth direction, are parallel. This latter structure gives origin to the sharp rocking peak, whereas the former one to the broad peak.

Fig. 7 Model of the columnar film growth in the V/Ni superlattice. The column axes are slightly tilted, and the averaged modulation is perpendicular to the substrate surface.

(a)

(b)

Fig. 8 (a) Columnar structure with modulation periodicity Λ , (b) Fourier transform of the columnar structure in k -space.

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