Comment on “Control of epitaxial growth for $SrBi_2Ta_2O_9$ thin films” [Appl. Phys. Lett. 72, 665 (1998)]

J. Lettieri, C. I. Weber, and D. G. Schlom

Citation: **73**, (1998); doi: 10.1063/1.122561
View online: http://dx.doi.org/10.1063/1.122561
View Table of Contents: http://aip.scitation.org/toc/apl/73/14
Published by the American Institute of Physics

COMMENTS

Comment on "Control of epitaxial growth for $SrBi_2Ta_2O_9$ thin films" [Appl. Phys. Lett. 72, 665 (1998)]

J. Lettieri, C. I. Weber, and D. G. Schlom
Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802-5005

(Received 24 March 1998; accepted for publication 4 August 1998)

[S0003-6951(98)01940-8]

Recently, Cho *et al.*[1] reported the epitaxial growth of *a*- and *c*-axis $SrBi_2Ta_2O_9$ films. While we agree with their analysis of the *c*-axis films, we believe that the x-ray patterns that they attributed to the growth of *a*-axis $SrBi_2Ta_2O_9$ films are actually due to an epitaxial impurity phase: δ-Bi_2O_3, β-Bi_2O_3, or $Bi_{7.80}Ta_{0.20}O_{12.20}$.[2] All three have nearly degenerate peaks in 2θ, χ, and ϕ with each other and would give rise to x-ray patterns consistent in both peak positions and peak intensities with those shown by Cho *et al.*,[1] which they attributed to *a*-axis $SrBi_2Ta_2O_9$. Below, we describe how epitaxial δ-Bi_2O_3 would give rise to the same x-ray diffraction patterns reported by Cho *et al.*[1] Similar arguments can be made for β-Bi_2O_3 and $Bi_{7.80}Ta_{0.20}O_{12.20}$ (see Table I).

In studying the epitaxial growth of $SrBi_2Ta_2O_9$ films,[3] we observed very similar θ–2θ x-ray diffraction patterns to those reported by Cho *et al.*[1] for films grown on $LaAlO_3(100)$ substrates by pulsed laser deposition at substrate temperatures of 750–800 °C and oxygen/ozone (~5%–10% ozone) pressures of 20–150 mTorr. An example is shown in Fig. 1. This θ–2θ plot alone is inconclusive for

TABLE I. Degenerate peaks for $SrBi_2Ta_2O_9$ and three impurity phases.[a]

Phase	Peaks	2θ (degrees)	χ[b] (degrees)	ϕ (degrees)
$SrBi_2Ta_2O_9$ (100)-oriented	200	32.34	90	⋯
	400	67.68	90	⋯
	115	28.95	51.94	0
	$20\underline{10}$	49.08	42.10	45
δ-Bi_2O_3 (100)-oriented	$2\overline{00}$	32.28	90	⋯
	400	67.76	90	⋯
	111	27.94	35.26	0
	220	46.43	45.00	45
$\beta=Bi_2O_3$ (110)-oriented	220	32.68	90	⋯
	440	68.47	90	⋯
	201	27.94	35.64	±0.81[c]
	400	46.89	45.00	45
$Bi_{7.80}Ta_{0.20}O_{12.20}$ (110)-oriented	220	32.77	90	⋯
	440	68.68	90	⋯
	201	27.94	35.71	±0.96[c]
	400	47.03	45.00	45

[a]The values are based on Cu $K\alpha_1$ radiation, bulk lattice constants (Refs. 2 and 5) and $\phi=0°$ chosen to be parallel to the in-plane [001] direction of the (100) $LaAlO_3$ substrate.
[b]$\chi=90°$ is perpendicular to the plane of the substrate.
[c]Assuming degenerate epitaxy (Ref. 4).

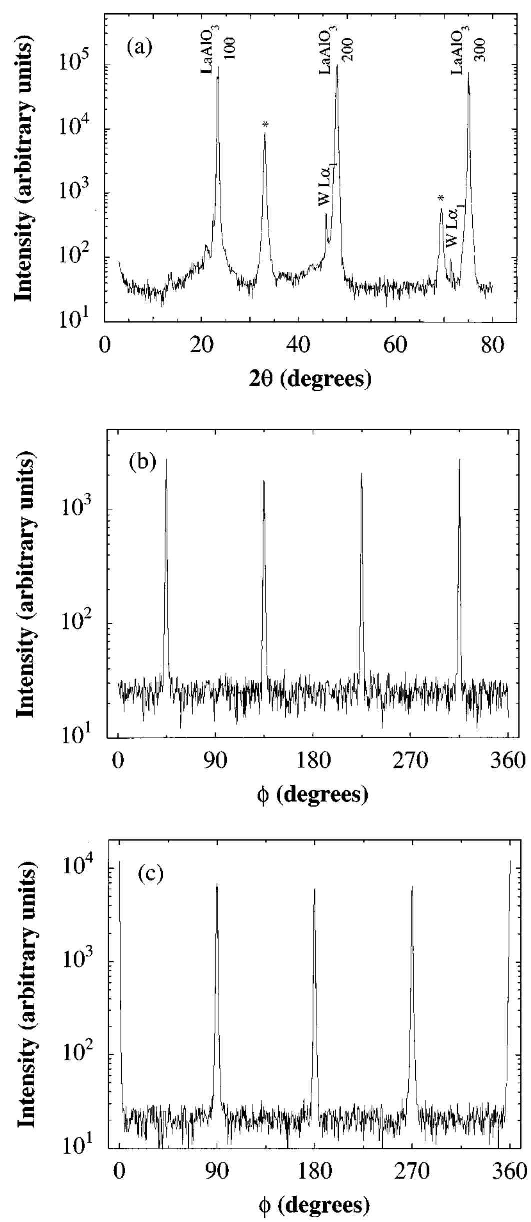

FIG. 1. X-ray diffraction patterns of a film grown under similar conditions as Cho *et al.* (Ref. 1) that does not contain $SrBi_2Ta_2O_9$. (a) θ–2θ (at $\chi=90°$) plot of an epitaxial impurity phase (labeled as *). (b) ϕ scan at $\chi=45.0°$ and $2\theta=47.5°$. (c) ϕ scan at $\chi=35.3°$ and $2\theta=28.5°$. These x-ray scans are all comparable to the scans presented in Figs. 1(a), 2(b), and 2(c) of Ref. 1, respectively.

phase determination, since the $h00$ δ-Bi_2O_3 peaks occur at nearly identical positions as the $SrBi_2Ta_2O_9$ $h00$ peaks (see Table I). Using four-circle x-ray diffraction we have found that the phase in our films has peaks consistent with one of the aforementioned degenerate impurity phases, but *inconsistent* with the growth of *a*-axis $SrBi_2Ta_2O_9$. We determined conclusively that our films do not contain *a*-axis $SrBi_2Ta_2O_9$ by conducting a ϕ scan of the 115 peak at $\chi \cong 54°$. No peaks were found, which indicates the absence of *a*-axis $SrBi_2Ta_2O_9$ (not shown).

The ϕ scans reported by Cho *et al.*[1] are insufficient to discriminate between the growth of epitaxial δ-Bi_2O_3 and $SrBi_2Ta_2O_9$. The 2010 reflection of *a*-axis $SrBi_2Ta_2O_9$ is indistinguishable from the 202 δ-Bi_2O_3 reflection in 2θ, χ, and ϕ [see Table I and Fig. 1(b)]. The final ϕ scan presented by Cho *et al.*,[1] a ϕ scan where $SrBi_2Ta_2O_9$ would yield 115 peaks, is capable of discriminating between the growth of *a*-axis $SrBi_2Ta_2O_9$ and δ-Bi_2O_3. Here, there is an overlap of peaks in 2θ and ϕ (if the orientation with the most favorable lattice match is considered), but in χ the peaks occur at complementary angles (see Table I). Since no χ values are reported by Cho *et al.*,[1] we can only speculate as to whether the reported ϕ scan might have been performed at the complementary χ angle. A plot showing our ϕ scan for our degenerate impurity phase is shown in Fig. 1(c).

The most compelling evidence, however, lies in a consideration of structure and lattice parameters. Cho *et al.*[1] see a fourfold symmetry in their ϕ scans, which they attribute to $SrBi_2Ta_2O_9$ growing with a "biepitaxial" structure. In fact, we believe that they see a fourfold symmetry (rather than a twofold symmetry as would be seen in a single domain, *a*-axis orthorhombic structure) because they are actually looking at the growth of (100)-oriented δ-Bi_2O_3, a cubic material. Finally, Cho *et al.*[1] note the lattice parameters that they determine from their x-ray measurements of "*a*-axis-oriented $SrBi_2Ta_2O_9$ films" ($a \approx b = 5.361$ Å, $c = 26.83$ Å) differ greatly from those observed for bulk $SrBi_2Ta_2O_9$ ($a = 5.5306$ Å, $b = 5.5344$ Å, $c = 24.9839$ Å),[5] or for their *c*-axis oriented $SrBi_2Ta_2O_9$ films ($a \approx b = 5.574$ Å, $c = 25.07$ Å). Although these highly distorted lattice constants appear unusual when indexed as $SrBi_2Ta_2O_9$, the c/a lattice parameter quotient is integral (5), as would be expected for a cubic phase that has been misindexed, i.e., δ-Bi_2O_3. We believe what they are classifying as one unit cell of $SrBi_2Ta_2O_9$ is really five unit cells of δ-Bi_2O_3. Similar arguments can be made for the other nearly degenerate impurities in place of δ-Bi_2O_3 in the above discussion of structure and lattice parameter.

[1] J. H. Cho, S. H. Bang, J. Y. Son, and Q. X. Jia, Appl. Phys. Lett. **72**, 665 (1998).
[2] Powder Diffraction File (International Center for Diffraction Data, Swarthmore, PA, 1995) JCPDS cards 27-52, 27-50, and 43-451.
[3] J. Lettieri, Y. Jia, M. Urbanik, C. I. Weber, J.-P. Maria, R. Uecker, P. Reiche, and D. G. Schlom, Presented at the 10th International Symposium on Integrated Ferroelectrics, Monterey, CA, 1998 (unpublished).
[4] S.-W. Chan, J. Phys. Chem. Solids **55**, 1137 (1994).
[5] A. D. Rae, J. G. Thompson, and R. L. Withers, Acta Crystallogr., Sect. B: Struct. Sci. **B48**, 418 (1992).