High optical quality ZnO epilayers grown on sapphire substrates by reactive magnetron sputtering of zinc target

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Abstract

Zinc oxide (ZnO) epilayers were grown on (0 0 0 1) sapphire substrates by reactive sputtering of a zinc target at substrate temperatures of 300 and 600 °C. High-resolution X-ray diffraction, UV–visible and photoluminescence (PL) measurements were carried out to obtain information about epitaxy, microstructure and optically active defects in these epilayers. Though the epilayer deposited at 600 °C showed a slightly smaller crystallite size along the growth direction as compared to that deposited at 300 °C, it was much superior in terms of other micro-structural parameters. It exhibited significantly small values of micro-strain (2 $\times$ $10^{-4}$), rocking curve width (~0.13°), mosaic twist (0.35°), and screw (6.6 $\times$ $10^{8}$ cm$^{-2}$)- and edge (2.9 $\times$ $10^{11}$ cm$^{-2}$)-type dislocation densities. Absorption and PL studies showed the high optical quality of the ZnO epilayer deposited at 600 °C, which exhibited a narrow (full-width at half-maximum—FWHM—96 meV) and intense band edge luminescence at room temperature. The micro-structural parameters and the sharp PL peak show that the reactively sputtered ZnO epilayer grown at 600 °C is comparable in epitaxial and optical quality with ZnO grown by other epitaxial processes.

Keywords:
A1. High-resolution X-ray diffraction
A3. rf Sputtering
B1. Zinc oxide
B2. Optical properties

1. Introduction

Zinc oxide (ZnO) is a wide-band-gap semiconductor with large exciton binding energy (60 meV), making it one of the most potential materials for realization of the next generation optoelectronic devices, operating in the short-wavelength region [1,2]. The electrical, piezoelectric, pyro-electric and ferroelectric properties of ZnO, along with its high chemical and thermal stability, and abundance, make it a very versatile material. It has several advantages over GaN, which is an established semiconductor for short-wavelength optoelectronic devices. These include the availability of simple crystal-growth technology and reasonably high-quality ZnO bulk single crystals. The high transparency of ZnO in the visible region makes it attractive for the development of transparent electronics, which can be integrated with ZnO-based optoelectronic devices and sensors [3]. Several growth techniques, such as spray pyrolysis, sputtering, pulsed laser deposition (PLD), molecular beam epitaxy (MBE) and metal–organic chemical vapour deposition (MOCVD), have been extensively used for the deposition of ZnO films and most of this work has been extensively reviewed [1–4]. High-quality ZnO epilayers have usually been grown on crystalline substrates, such as sapphire by PLD [5–7], MBE [8–10] and MOCVD [11,12]. In recent years, there has been an increasing interest in the growth of ZnO epilayers by sputtering [13–22]. This is owing to the advantages of sputtering in terms of versatility, large-area deposition, low cost, high deposition rates and scalability. In most cases, ZnO films have been deposited by sputtering of a ZnO ceramic target. The ceramic targets are mechanically fragile and expensive, and yield lower deposition rates compared to metallic targets. The composition of the target also has a strong effect on stoichiometry of films, often leading to problems of reproducibility. In comparison, reactive sputtering of a metallic zinc target has been less frequently used to deposit ZnO films and more specifically, the growth of epitaxial ZnO films by this approach has been rarely reported [15]. The reactive sputtering process is relatively simple and attractive because the stoichiometry of the films is controlled only by oxygen partial pressure during sputtering and deposition rates are...
also relatively higher. In this background, a detailed study of epitaxial ZnO films deposited by reactive rf sputtering of a metallic zinc target assumes significance.

In a recent work [23] on polycrystalline ZnO films deposited on quartz substrates by reactive rf magnetron sputtering of metallic zinc target, it has been reported that the film deposited at 300 °C showed a strong and nearly complete c-axis orientation of crystallites. The films deposited at higher substrate temperatures showed negligible uniform strain but a steady increase in the misorientation of the crystallites with respect to the film surface. It was however found that the ZnO film deposited at 600 °C exhibited the sharpest absorption edge and a strong and narrow room temperature band edge photoluminescence (PL). In view of the excellent structural and optical properties of ZnO films deposited by reactive sputtering of a zinc target, this approach has been used in the present work to grow ZnO epilayers on sapphire substrates. The epilayers have been grown at substrate temperatures of 300 and 600 °C and a comprehensive micro-structural investigation has been carried out by high-resolution X-ray diffraction (HRXRD) measurements to assess their micro-structural parameters. The optical properties of the epilayers have been studied by UV–visible absorption and PL measurements. The present work has shown that ZnO epilayers can be grown by reactive sputtering of a zinc target at substrate temperatures of 300–600 °C. Moreover, the film grown at 600 °C exhibits high epitaxial and optical quality, not reported earlier using this approach and comparable to the best reported characteristics of ZnO epilayers grown on sapphire substrates by any technique.

2. Experimental details

ZnO epilayers were deposited on sapphire substrates by reactive rf magnetron sputtering. A 99.9%-pure Zn target of 3-in diameter was used. The target to substrate distance was 55 mm. The base pressure was 1 × 10⁻⁷ mbar. The flow rates of argon (24 SCCM) and oxygen (6 SCCM) were controlled by mass flow controllers and the oxygen to argon ratio was kept constant at 20%. The deposition was carried out at a working pressure of 10⁻² mbar, after pre-sputtering with argon for 10 min. The sputtering power was maintained at 400 W during deposition. The depositions were carried out on sapphire substrates at temperatures of 300 and 600 °C. The thickness of the ZnO epilayers studied in this work was 24–600 nm.

HRXRD measurements were carried out in omega (ω)-, ω–2ω- and ϕ-scan geometries using PANalytical X’Pert MRD system. The incident beam optics had a four-bounce hybrid monochromator, which ensured Cu Kα1 (1.54056 Å) output collimated to about 20 arcsec in the plane of scattering. A ½ slit was placed at the output before the detector. The reciprocal lattice maps were recorded by placing a three-bounce Ge(2 2 0) collimator before the detector and recording a series of ω- and ω-2ω-scans. Absorption studies were carried out on Perkin-Elmer Lambda-950 UV–visible spectrophotometer. PL measurements were carried out at room temperature using a 325 nm He–Cd laser and a JOBIN YVON HR-460 monochromator.

3. Results and discussion

Phi (ϕ) scans of asymmetric (1 0 1 1) reflections were carried out on ZnO epilayers grown on sapphire substrates at substrate temperatures of 300 and 600 °C. The ϕ-scans of (1 0 1 1) reflection from the ZnO epilayers along with the ϕ-scans of (1 0 1 4) reflections of the sapphire substrate are shown in Fig. 1. Six peaks in the ϕ-scan reveal six-fold symmetry of the hexagonal ZnO lattice, which shows the presence of in-plane orientation of crystallites and thus the evidence for epitaxial growth of ZnO on sapphire substrate. Comparison of ϕ-scans of ZnO epilayers with sapphire substrates shows that in both cases, the ZnO lattice is rotated by 30° with respect to the sapphire lattice. This shows that the ZnO lattice (a = 3.25 Å) aligns with oxygen sub-lattice (lattice constant 2.75 Å) and not with the aluminium sub-lattice (lattice constant 4.75 Å), implying a lattice mismatch of ~18%.
Under these conditions a zinc atom is bonded to one oxygen atom in $\text{Al}_2\text{O}_3$, and the in-plane epitaxial relationship is $\text{ZnO} \parallel \alpha-\text{Al}_2\text{O}_3$ [1 1 2 0] [8,24]. The 30° rotation of ZnO lattice with respect to sapphire at temperatures as low as 300 °C is in contrast with the occurrence of epitaxial growth of ZnO at relatively higher temperatures, in the case of deposition techniques such as PLD, MOCVD and MBE [7,25,26]. It has been widely reported [25] that ZnO epilayers show 30° rotation of ZnO with respect to sapphire lattice, and hence a reduced lattice mismatch, usually at higher growth temperatures, which has been attributed to the increased surface mobility of ad atoms. However, ZnO epilayers grown on c-sapphire by reactive sputtering have been reported [15] to maintain the epitaxial relationship $\text{ZnO} \parallel \alpha-\text{Al}_2\text{O}_3$ [1 1 2 0], even at substrates temperatures down to 80 °C. The present results agree with the observations made in Ref. [15] and thus support the corresponding inference [15] that the low-temperature epitaxy of reactively sputtered ZnO can be explained by the presence of energetic species such as zinc ad atoms, which possess high surface mobility in low-oxygen conditions, and thus make the epitaxial process less dependant on substrate temperature.

3.1. Micro-structural studies

The micro-structure and epitaxial quality of ZnO grown on sapphire substrates at 300 and 600 °C have been investigated by high-resolution omega (ω)- and ω–2θ-scans. Epitaxial films grown on lattice-mismatched substrates can be considered to consist of oriented mosaic blocks or single crystallites, with certain mean vertical (along growth direction) and lateral (along plane of growth) dimensions. The mosaic blocks can be slightly mis-oriented with respect to each other, and the mosaic tilt and twist, respectively, refer to the mis-orientation of the mosaic blocks out of the sample plane and within the sample plane. The presence of defects dilates/contracts the lattice close to it, resulting in a local strain close to the defect, which is referred to as the micro-strain. The finite crystallite size, tilt, twist and micro-strain cause broadening (Δq) of (0 0 0 l) reflections in the ω–2θ-scans, which can be, respectively, estimated from the intercept and slope of the linear Williamson–Hall plots [28]. The finite vertical coherence length and micro-strain cause a broadening (Δql) of (0 0 0 l) reflections in the ω–2θ-scans, which can be, respectively, estimated from the intercept and slope of the linear Williamson–Hall plots of Δql (FWHM along $q_l$ direction) vs. $q_l$. Here, $q_l$ is the magnitude of the position of (0 0 0 l) point in the reciprocal space. Similarly, the finite lateral coherence length and mosaic tilt cause broadening (Δqs) in the $q_s-q_s$ plane of (0 0 0 l) reflections in ω-scans, which can be estimated, respectively, from the intercept and slope of the linear Williamson–Hall plots of Δqs (FWHM along $q_s$ direction) vs. $q_s$. The in-plane rotation of mosaic blocks or mosaic twist can be estimated from the FWHM of the reflection from a plane, which is suitably oriented with respect to the (0 0 0 2) plane.

The 2θ-values corresponding to (0 0 0 2) reflection were 34.36° and 34.43° for the ZnO epilayers deposited at 300 and 600 °C, respectively. These values are close to the corresponding JCPDS value of 34.439° (File no. 05-664), thus indicating the absence of uniform strain in the epilayers, as reported [23] earlier for the polycrystalline ZnO films deposited on quartz substrates; ω–2θ-scans for the symmetric (0 0 0 2), (0 0 0 4) and (0 0 0 6) reflections for ZnO epilayers deposited at substrate temperatures of 300 and 600 °C are shown in Fig. 2(a). For all cases, the FWHM values in reciprocal space (Δq) are also indicated. In the case of epilayer deposited at 300 °C, the width of the ω–2θ peak increases significantly with order of reflection, while the corresponding increase is much less in the epilayer deposited at 600 °C. The micro-strain and vertical coherence length (crystallite size along growth direction) were estimated from the Williamson–Hall plots (Δqs vs. $q_s$) shown in Fig. 2(b), and the values of these parameters are listed in Table 1. The micro-strain in the epilayer deposited at 600 °C is $2.0 \times 10^{-4}$, which is much smaller than the corresponding value of $1.0 \times 10^{-3}$ in the epilayer deposited at 300 °C. It is however found that the vertical coherence length (average crystallite size along growth direction) of the epilayer deposited at 600 °C (~85 nm) is much smaller than that of the epilayer deposited at 300 °C (~145 nm).

Fig. 3(a) shows the ω-scans for symmetric (0 0 0 2), (0 0 0 4) and (0 0 0 6) reflections of the ZnO epilayers. For all the cases, the
FWHM values in reciprocal space (Δqₓ) are also indicated. The ω-scans of the epilayer deposited at 600 °C were found to be much narrower compared to the epilayer deposited at 300 °C.

![Fig. 3.](image)

**Table 1**

<table>
<thead>
<tr>
<th>Substrate temp. (°C)</th>
<th>Lateral coherence length (nm)</th>
<th>Mosaic tilt (deg)</th>
<th>Vertical coherence length (nm)</th>
<th>Micro-strain (× 10⁻³)</th>
<th>Mosaic twist (deg)</th>
<th>Screw dislocation (cm⁻²)</th>
<th>Edge dislocation (cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>1600 ± 300</td>
<td>0.75</td>
<td>145 ± 55</td>
<td>1.0</td>
<td>1.75</td>
<td>1.5 × 10¹⁰</td>
<td>7.3 × 10¹²</td>
</tr>
<tr>
<td>600</td>
<td>&gt; Limit of measurements</td>
<td>0.16</td>
<td>85 ± 5</td>
<td>0.2</td>
<td>0.35</td>
<td>6.6 × 10⁸</td>
<td>2.9 × 10¹¹</td>
</tr>
</tbody>
</table>

The corresponding Williamson–Hall plots of Δqₓ vs. q are shown in Fig. 3(b) for both the epilayers. The mosaic tilt and lateral coherence length (average in-plane crystallite size) were estimated from the slope and intercept of the plots, and are listed in Table 1. Though linear fits of Δqₓ vs. q plots for the two epilayers showed significantly different slopes, the corresponding intercepts are nearly zero, indicating large lateral dimensions. The average value of mosaic tilt of 0.16° for the epilayer deposited at 600 °C is significantly smaller than the value of 0.75° obtained for the epilayer deposited at 300 °C.

Another important micro-structural parameter representing the quality of epitaxy is the mosaic twist, which is a measure of average in-plane rotation of mosaic blocks. It has been earlier reported [5] that the value of twist can be evaluated using a series of reflections and by fitting a model that describes the effect of tilt and twist on rocking curve widths of various reflections. It has however been shown [29] that the width of (1 0 1 1) reflection gives a good measure of the value of twist. Thus, in the present work, ω-scans of (1 0 1 1) reflection of the ZnO epilayers shown in Fig. 4 were used to estimate the mosaic twist. The epilayer deposited at 600 °C showed a very narrow peak with a FWHM value of 0.35° as compared to 1.75° for the epilayer deposited at 300 °C. It may be noted that the extremely small in-plane twist of 0.35°, seen in the epilayer deposited at 600 °C, is much lesser than the typically reported values of mosaic twist in ZnO epilayers deposited on c-sapphire by MBE [9] (0.8°) and PLD [5] (1°–1.5°). The value of twist measured in the present case is in fact quite comparable to values obtained in MBE-grown ZnO epilayers on lattice-matched substrates, such as GaN/sapphire [29] (0.25°) and MgO/sapphire [30] (0.34°).

The HRXRD data of (0 0 0 2) and (1 0 1 1) reflections have been used to evaluate the dislocation density in ZnO epilayers. TEM studies [5,29] have shown that in wurtzite systems, the dominant types of dislocations present in the layers are of pure-screw type and pure-edge type, though mixed dislocations with relatively lower concentrations have also been observed.
The pure-screw-type dislocations are along [0 0 0 1] directions with Burger’s vector \( \langle 0 0 0 1 \rangle \). The pure-edge-type dislocations are also along [0 0 0 1] directions but with Burger’s vector (1/3) \\
\( \langle 1 1 \bar{2} 0 \rangle \). Considering random distribution of these dislocations, the density of edge and screw dislocations can be evaluated following Refs. \([31,32]\). The values of dislocation density of the edge and screw types are listed in Table 1. The results show that the epilayer deposited at 600 °C shows much smaller (by \( \sim \)2 orders of magnitude) densities of both edge-type and screw-type dislocations, as compared to the epilayer deposited at 300 °C. Edge-type dislocation densities are found to dominate in both the cases, which agrees well with the reported results for MBE-grown ZnO \([10,29]\) and MOCVD-grown GaN \([28]\) epilayers.

Various techniques, such as X-ray diffraction (XRD), TEM and AFM have been used to calculate the dislocation densities of ZnO and GaN epilayers \([28,29,31,33]\). Though TEM is the most commonly used technique, it has been shown that there exists a good agreement between the results obtained from XRD and TEM data \([28,29]\). Thus the values of screw \((6.6 \times 10^{10} \text{ cm}^{-2})\) and edge \((2.9 \times 10^{11} \text{ cm}^{-2})\) dislocation density obtained for the ZnO epilayer deposited at 600 °C have been compared with reported values of dislocation densities obtained by XRD and TEM studies. The density of screw dislocations obtained in the present work is quite comparable to the reported values for ZnO epilayers grown on MgO/sapphire by MBE \([29]\) \((2.4 \times 10^{10} \text{ cm}^{-2})\) by XRD and on sapphire by PLD \([34]\) \((2.5 \times 10^{10} \text{ cm}^{-2})\) by TEM. However, the density of edge dislocations is about an order of magnitude higher than the reported values for ZnO epilayers grown by MBE \([29,35]\) \((0.5–1 \times 10^{10} \text{ cm}^{-2})\) and by PLD \([34]\) \((1.6 \times 10^{10} \text{ cm}^{-2})\). It may be mentioned that dislocation densities for sputtered ZnO epilayers have not been reported earlier.

A distinct asymmetry is seen in the lower intensity region of the \(\omega-2\theta\) peaks (Fig. 2(a)) in all the cases. This is more clearly seen in the higher order reflections. Interestingly, the asymmetry lies on the higher \(2\theta\) side for the epilayer deposited at 300 °C but shifts to the lower \(2\theta\) side for the epilayer deposited at 600 °C. The asymmetry on the higher \(2\theta\) side in epilayers deposited at 300 °C implies reduced \(d\)-values, which may be caused by a decrease in lattice parameter in the vicinity of point defects such as oxygen vacancies. This is consistent with the larger micro-strain present in the epilayers deposited at 300 °C, which is also an indication of the significant presence of point defects. The drastic decrease in micro-strain of the epilayer deposited at 600 °C and absence of asymmetry on higher \(2\theta\) side is an evidence of the reduction of point defects in this case. The consequent narrowing of \(\omega-2\theta\)-scans is however seen to result in the appearance of a small asymmetry, which is now on the lower \(2\theta\) side. This asymmetry could have been masked by the large width of \(\omega-2\theta\) curves in the case of the film deposited at 300 °C. The asymmetry on the lower \(2\theta\) side can be attributed to two possible reasons. It could result from the presence of a strained interfacial layer having a larger ‘\(c\)’ and smaller ‘\(a\)’ value (in-plane biaxial compressive strain). As the \(\phi\)-scans of the epilayers have shown that the ZnO lattice aligns with oxygen sub-lattice of sapphire, which has a smaller lattice constant of 2.75 Å compared to 3.25 Å for ZnO, the oxygen sub-lattice could cause the reduction in ‘\(a\)’ parameter of ZnO at the ZnO/sapphire interface along with a corresponding increase in the ‘\(c\)’ parameter. Another possible reason for the presence of asymmetry on the lower \(2\theta\) side, which can become important at higher substrate temperatures, is the mismatch in thermal expansion coefficients of ZnO and sapphire. The lattice strain caused by thermal expansion coefficient mismatch is given by

\[
\varepsilon_{th} = \left( x_{\text{ZnO}} - x_{\text{sapphire}} \right) (T_s - T_0)
\]

where \(x_{\text{ZnO}}\) and \(x_{\text{sapphire}}\) are the thermal expansion coefficients for ZnO and sapphire, and \(T_s\) and \(T_0\) are the deposition temperature and room temperature, respectively. Using values of \(x_{\text{ZnO}} \approx 3 \times 10^{-6} \text{ K}^{-1}\) and \(x_{\text{sapphire}} \approx 7.5 \times 10^{-6} \text{ K}^{-1}\) along \(a\)-axis, it can be seen that the compressive thermal strain in the epilayer deposited at 600 °C is nearly twice that in the epilayer deposited at 300 °C. This compressive strain in the growth plane may also cause elongation of \(c\)-axis, as seen in the present case.

It is thus inferred from above that though there is a decrease of micro-strain in the epilayer deposited at 600 °C compared to that deposited at 300 °C, the biaxial compressive strain caused by the substrate is noticeable only in the former case. This may be because the broadening due to micro-strain is much reduced in this case and possibly, there is also an increase in the magnitude of biaxial compressive strain at higher substrate temperatures.

The above conjecture of the presence of biaxial strain in the ZnO epilayer grown at 600 °C has been confirmed from the reciprocal lattice maps for \((0 0 0 2)\) and \((1 0 1 1)\) reflections. These maps are shown in Figs. 5(a) and (b). An increase in the lattice parameter ‘\(c\)’ results in a reduced reciprocal lattice vector \(c\). This shows up as a distinct asymmetry on the lower \(Q_z\) side. Under biaxial strain, an increase in ‘\(c\)’ results in a decrease in the value of ‘\(a\)’. This decrease in the lattice parameter ‘\(a\)’ results in a corresponding increase in reciprocal lattice vectors \(\bar{A}\) and \(\bar{B}\), which results in a distinct asymmetry of the \((1 0 1 1)\) reciprocal map on the higher \(Q_x\) side, as seen in Fig. 5(b). These features of the reciprocal maps confirm the presence of a biaxially strained layer of ZnO, most likely at the ZnO/sapphire interface, which could originate from one of the two reasons mentioned above.

### 3.2. Optical studies

Optical properties of the ZnO epilayers deposited at 300 and 600 °C were studied using UV–visible absorption and PL measurements. The absorption coefficient \((x)\) was estimated from specular
reflectance \((R)\) and transmittance \((T)\) data, and thickness \((d)\) of the epilayers, using the approximate expression

\[
T(\lambda) \approx (1 - R(\lambda))^2 e^{-2\pi d/\lambda}
\]

for a self-supporting film, with high absorption and low reflectance. The corresponding plots of \(x^2\) vs. \(h\nu\) are shown in Fig. 6, from which the direct band gap of the epilayers has been obtained. The epilayers deposited at 300 and 600°C exhibit nearly equal values of band gap \(\approx 3.3\) eV, which is close to the bulk band gap of ZnO. It is however seen that the epilayer deposited at 600°C shows a significantly sharper \(x^2\) vs. \(h\nu\) plot and a much reduced sub-bandgap absorption, compared to the epilayer deposited at 300°C, indicating a significant reduction in optically active defects and disorder with increase of substrate temperature.

PL measurements were carried on ZnO epilayers at room temperature. The PL spectra of epilayers deposited at substrate temperatures of 300 and 600°C are shown in Fig. 7. Both the epilayers show strong emission at \(\sim 376\) nm, attributed to the near band edge UV luminescence of ZnO. In the case of the epilayer deposited at 300°C, weak and broad bands are also seen in green–yellow region, with intensity comparable to the band edge emission. This luminescence is known to arise from oxygen vacancies and zinc interstitials in ZnO [36,37]. In contrast, the epilayer deposited at 600°C shows a very high intensity of band edge luminescence, with comparatively negligible defect luminescence. The half-width of the band edge peak for the epilayer deposited at 600°C is \(\sim 96\) meV, which is significantly smaller than the value of \(\sim 157\) meV for the epilayer deposited at 300°C. The FWHM of \(\sim 96\) meV for the room temperature band edge PL peak is quite comparable to the corresponding results reported for high-quality ZnO epilayers grown by sputtering [13,14,17], PLD [38], MOCVD [39] and MBE [8] techniques. As typical examples, PL width values of 117 meV [8] and 106 meV [16] have been reported, for MBE-grown ZnO epilayers (with \(0 0 0 2\) rocking curve width \(\sim 0.005°\)) and homo-epitaxial ZnO grown by magnetron sputtering (with \(0 0 0 2\) rocking curve width \(\sim 10\) arcsec), respectively. The PL results are thus in excellent agreement with absorption studies and reiterate that oxygen-deficiency-related point defects in these epilayers decrease with an increase in substrate temperature. The reduction in point defects correlates well with the significant decrease in micro-strain to a very low value \(\sim 2 \times 10^{-4}\) in the epilayer deposited at 600°C.

4. Conclusions

HRXRD studies have shown that ZnO epilayers deposited on sapphire substrates by reactive rf magnetron sputtering of a zinc target, grow epitaxially at 300°C as well as 600°C. Both the epilayers show large lateral crystallite size (\(\sim 1000\) nm). Though the epilayer deposited at 600°C has a slightly smaller crystallite size along growth direction (\(\sim 85\) nm) compared to the epilayer deposited at 300°C (\(\sim 145\) nm), it is much superior in terms of all other micro-structural parameters and optical quality. Most importantly, the micro-strain in the epilayer deposited at 600°C is \(\sim 2 \times 10^{-4}\), as compared to the value of \(\sim 1 \times 10^{-3}\) in the epilayer deposited at 300°C. It may be noted that the micro-strain of \(2 \times 10^{-4}\) and the mosaic twist of \(0.35°\) are extremely small values, which have not been commonly reported for epitaxially grown ZnO. The \((0 0 0 2)\) rocking curve width of 0.13° is also comparable to the best reported results for sputtered [13,14,17], PLD-grown [5,6,38] and MOCVD-grown [11,39] ZnO, though it is higher than the rocking curve widths measured in MBE-grown epilayers [8,9]. The epilayer deposited at 600°C shows a low density \(\sim 6.6 \times 10^6\) cm\(^{-2}\) of screw dislocations, while the density \(\sim 2.9 \times 10^{11}\) cm\(^{-2}\) of edge dislocations is about an order of magnitude higher than the reported values for ZnO epilayers grown by MBE and PLD. The epilayer also exhibits the presence of a biaxially strained interface layer at the ZnO/sapphire interface caused by mismatch of either lattice or thermal expansion coefficients.

The FWHM of \(\sim 96\) meV for the band edge PL peak at room temperature seen in the epilayer deposited at 600°C is quite comparable to the results obtained on high-quality epitaxial ZnO grown by a variety of techniques. It is thus inferred that the ZnO deposited on sapphire by reactive rf magnetron sputtering of a metallic zinc target exhibits high epitaxial and optical quality not reported earlier. This is attributed to its superior micro-structural parameters, especially the extremely low values of micro-strain and mosaic twist that can be achieved at a moderate substrate temperature of 600°C.

Acknowledgements

The financial support from MHRD (Government of India) for this work is gratefully acknowledged. Sukhvinder Singh is thankful to CSIR, New Delhi (India) for Senior Research Fellowship. Tapas

Fig. 6. The \(x^2\) vs. \(h\nu\) plots for ZnO epilayers deposited at substrate temperatures of 300 and 600°C.

Fig. 7. PL spectra of ZnO epilayers deposited at substrate temperatures of 300 and 600°C.
Ganguli and Ravi Kumar are thankful to Dr. S.M. Oak for support and encouragement.

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