▼ Journal of Research (Science), Bahauddin Zakariya University, Multan, Pakistan.
Vol. 17, No. 2, April 2006, pp. 137-144 ISSN 1021-1012

OPTICAL BAND GAP OF ZnO THIN FILMS DEPOSITED BY ELECTRON BEAM EVAPORATION

M. Y. Nadeem, S. Liaqat Ali, M. F. Wasiq, A. M. Rana
Department of Physics, Bahauddin Zakariya University, Multan, Pakistan
Manzar Abbas
Pakistan Council of Renewable Energy Technologies, Islamabad

Abstract

Optical band gap of ZnO thin films deposited by electron beam evaporation at evaporation rates ranging 5 Ås^{-1} to 15 Ås^{-1} and thickness ranging 1000Å to 3000Å is presented. Deposited films were annealed at 573K for one and half hour. The variations in the optical band gap were observed and showed decreasing behavior from 3.15eV to 3.05 eV, from 3.18 eV to 3.10 eV and from 3.19 eV to 3.18 eV for films with respective thickness 1000Å, 2000 Å, 3000 Å on increasing the evaporation rate from 5 Ås^{-1} to 15 Ås^{-1} by keeping thickness constant.

Keywords: Optical absorption, optical band gap, ZnO thin films.

Transparent conducting oxides (TCOs) are utilized for a wide range of applications, such as optoelectronics, photocatalysts, photoelectrocatalysts, the flat panel display, gas sensors, the common and pixel electrode in thin film transistor liquid crystal display [Chae 2001] or the window layers for thin film solar cells based on Si [Sang *et al*. 1999] or Cu(InGa)Se_2 [Chaisitsal *et al*. 1999]. Although indium tin oxide (ITO) is very commonly used as TCO, research for an immediate alternative to ITO is desired because of the high cost and scarcity of the indium element. In recent years zinc oxide (ZnO) has attracted the attention of researchers because the Zn element is abundant. The lower production costs and easier processing for etching of ZnO than ITO are being expected.

Being semicoducting, photoconducting, piezoelectric and optical waveguide material [Ohgaki *et al*. 2003, Yukio *et al*. 2003, Wang 2004, Chuan-Lei 2005] ZnO has also found a wide range of scientific and technological applications. It exhibits a large band gap, fairly high refractive index [France 2004], a maximum solar transmittance of about 85% to 95% with a minimum resistivity as low as ~7x10^{-5} ohm-cm [Minami *et al*. 1985]. Pure ZnO is highly transparent in visible spectral region and highly reflective in IR region. ZnO thin films are also being used as a detector for oxidizing and reducing gases in chemical sensors, transparent electrodes in light emitting diodes [Chen *et al*.], T.V., Camera displays and heat mirrors.

ZnO thin films are being grown by various deposition methods such as sputtering [Agashe *et al*. 2003], molecular beam epitaxy (MBE) [Nakahara *et al*. 2001, Kato *et al*. 2002], metal organic chemical vapor deposition (MOCVD) [Gorla *et al*. 1999], pulsed laser deposition (PLD) [Myoung *et al*. 2002] and reactive plasma deposition (RPD) [Sakai *et al*. 2006].

In the present investigations, ZnO thin films were deposited by electron beam evaporation on glass substrates and characterized optically. Effects of the evaporation rate and thickness on the variation of band gap of these films have been discussed.

ZnO thin films were deposited by electron beam evaporation under a residual air pressure of 5×10^{-5} torr, with evaporation rates in the range 5 – 15 $Ås^{-1}$ and thickness varying as 1000 – 3000 Å using a Leybold–Heraeus A550V vacuum coating unit at Pakistan Council of Renewable Energy Technologies, Islamabad. Aluminum crucible was used for the evaporation of highly pure ZnO, while “Sail Brand No. 23, China” glass acted as substrate. Deposited films were annealed at 573 K for one and half hour and then characterized optically using Hitachi U-2001 UV/VIS/NIR double beam spectrophotometer at the Department of Physics, Bahauddin Zakariya University, Multan, Pakistan. Optical absorbance and transmittance of each of these thin film samples were recorded in the wavelength range 190–1100 nm at room temperature ~300K.

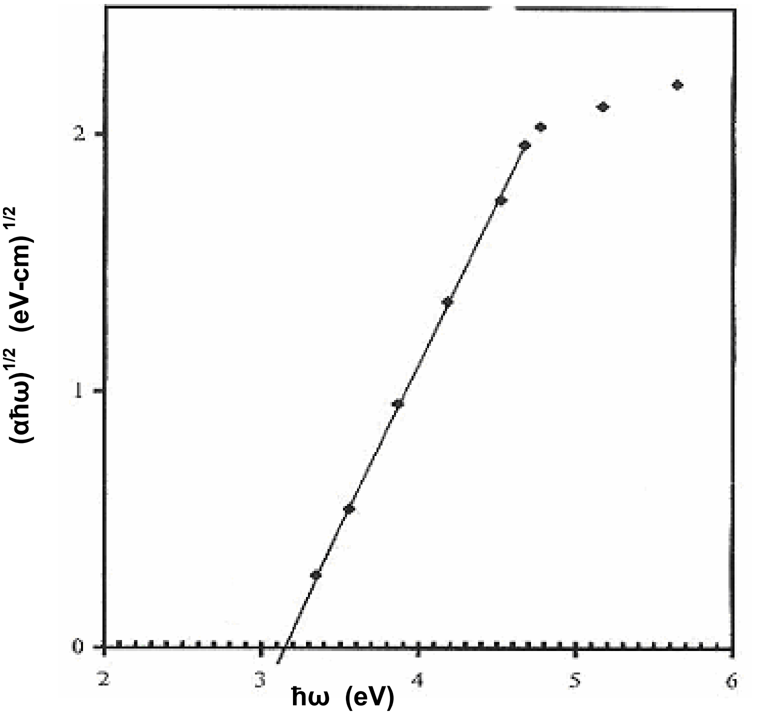

Fig. 1: Plot of $(\alpha \hbar \omega)^{1/2}$ versus photon energy $\hbar \omega$ to determine optical band gap of ZnO thin film (Thickness 1000Å, Evaporation Rate 5 $Ås^{-1}$).

Deposited films were highly transparent and free from pinholes. These thin films showed very good cohesion with the substrates. Optical absorbance spectra were analyzed to evaluate the optical band gap energy using a well-known relation [Berger and Pamplin 1993, Fallah *et al*. 2006]

$$\alpha(\omega) = \frac{A\,(\hbar\omega - E_{opt})^2}{\hbar\omega} \qquad (1)$$

Where α is the absorption coefficient, A the constant, $\hbar\omega$ the photon energy and E_{opt} the band gap energy. Eq. 1 was used to determine the band gap E_{opt} of the prepared samples. For this purpose, $(\alpha\hbar\omega)^{1/2}$ was plotted against the photon energy $\hbar\omega$. These plots are shown in Figs. 1-8 as a function of the evaporation rate as well as thickness. Values of E_{opt} for all samples are presented in Table 1 and lie in the range 3.05-3.19eV. It is obvious from Table 1 that band gap decreases on increasing the evaporation rate keeping thickness constant. A similar result was found by Fallah *et al*. [2006] for ITO films. A characteristic of the film that is influenced by its crystallinity is the optical energy gap. The more crystalline film, the higher the electron concentration, the higher the energy gap [Fallah *et al*. 2006]. Thus the above result can be explained by considering the fact that an increase in the evaporation rate results in an increase in the degree of disorder (showing less crystallinity) in the structure of deposited material. This introduces more localized states within the band gap and number of band tails also increase thus lowering the band gap.

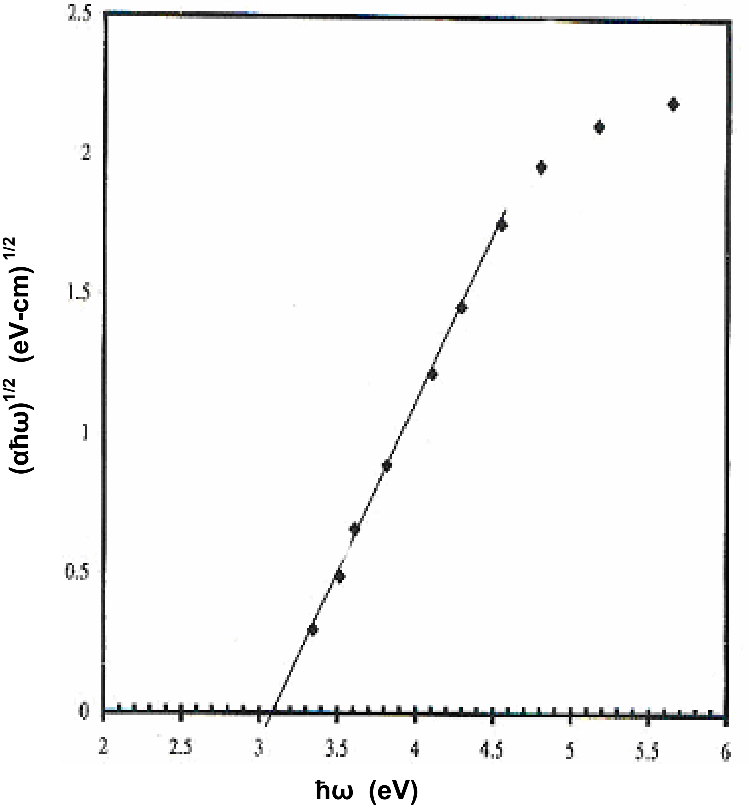

Fig. 2: Plot of $(\alpha\hbar\omega)^{1/2}$ versus photon energy $\hbar\omega$ to determine optical band gap of ZnO thin film (Thickness 1000Å, Evaporation Rate 10 Ås^{-1}).

The increasing band gap with an increase in thickness may be due to a rise in the grain size of ZnO thin films caused by annealing in air [Kashani 1998]. Table 1 also shows that the band gap increases with increasing thickness of these films by keeping evaporation rate constant [Srikant and Clarke 1997].

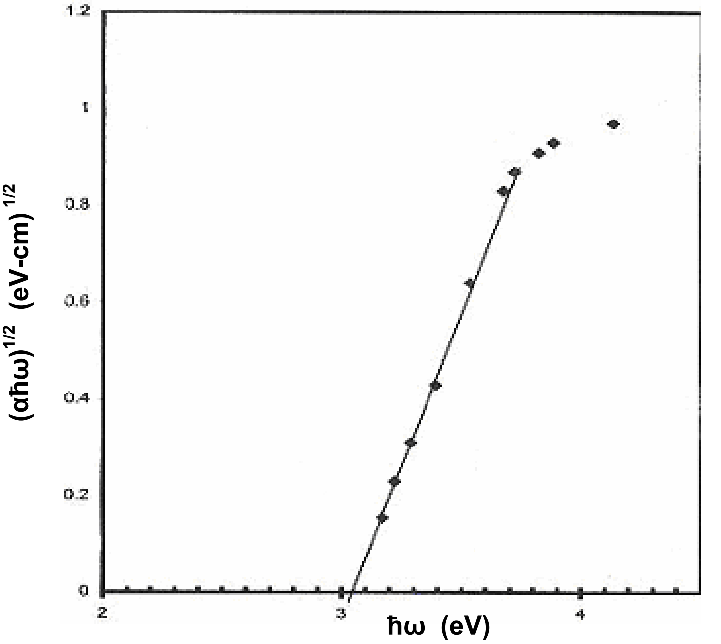

Fig. 3: Plot of $(\alpha\hbar\omega)^{1/2}$ versus photon energy $\hbar\omega$ to determine optical band gap of ZnO thin film (Thickness 1000Å, Evaporation Rate 15 Ås^{-1}).

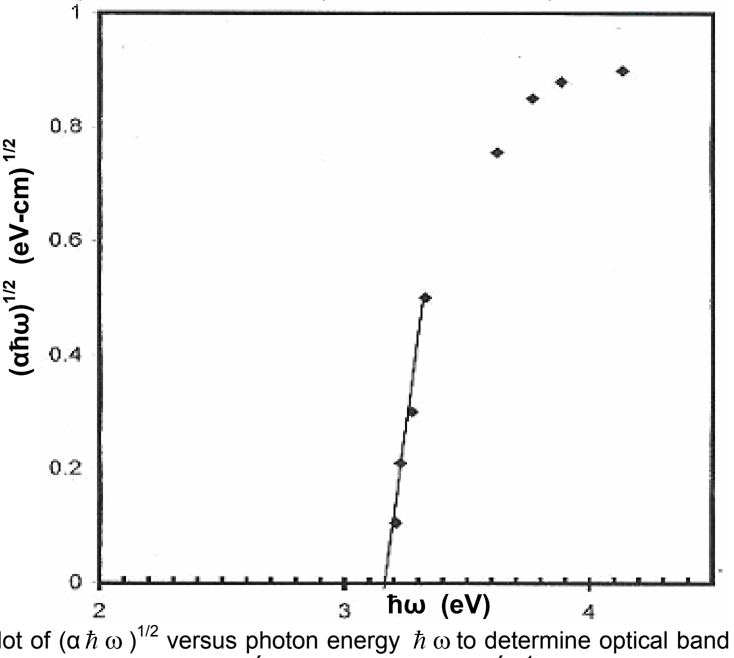

Fig. 4: Plot of $(\alpha\hbar\omega)^{1/2}$ versus photon energy $\hbar\omega$ to determine optical band gap of ZnO thin film (Thickness 2000Å, Evaporation Rate 5 Ås^{-1}).

Table 1: Optical band gap of ZnO thin films as a function of evaporation rate.

Thickness (Å)	Optical Band gap (eV)		
	Evaporation Rate (5 Ås^{-1})	Evaporation Rate (10 Ås^{-1})	Evaporation Rate (15 Ås^{-1})
1000	3.15	3.10	3.05
2000	3.18	3.14	3.10
3000	3.19	3.18	--

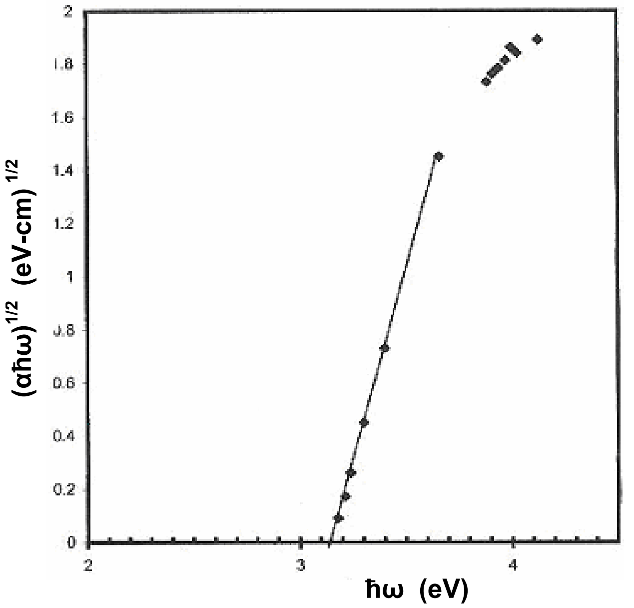

Fig. 5: Plot of $(\alpha\hbar\omega)^{1/2}$ versus photon energy $\hbar\omega$ to determine optical band gap of ZnO thin film (Thickness 2000Å, Evaporation Rate 10 Ås^{-1}).

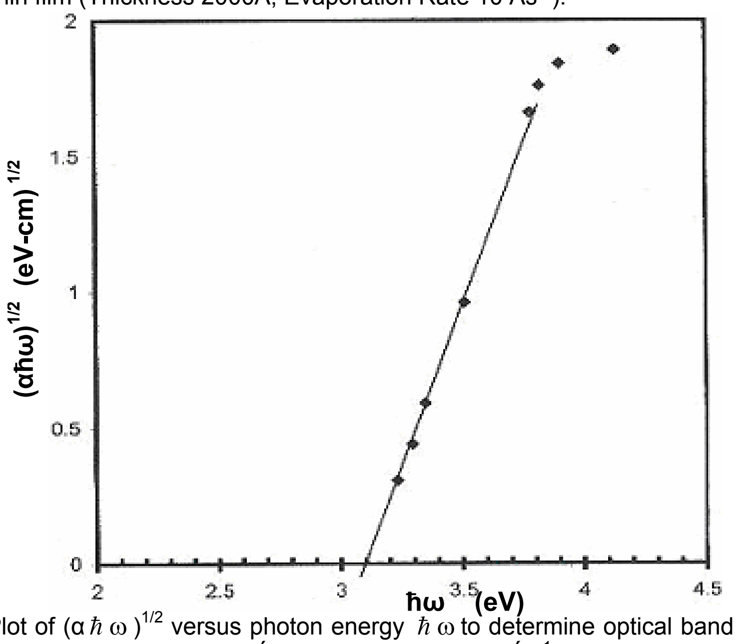

Fig. 6: Plot of $(\alpha\hbar\omega)^{1/2}$ versus photon energy $\hbar\omega$ to determine optical band gap of ZnO thin film (Thickness 2000Å, Evaporation Rate 15 Ås^{-1}).

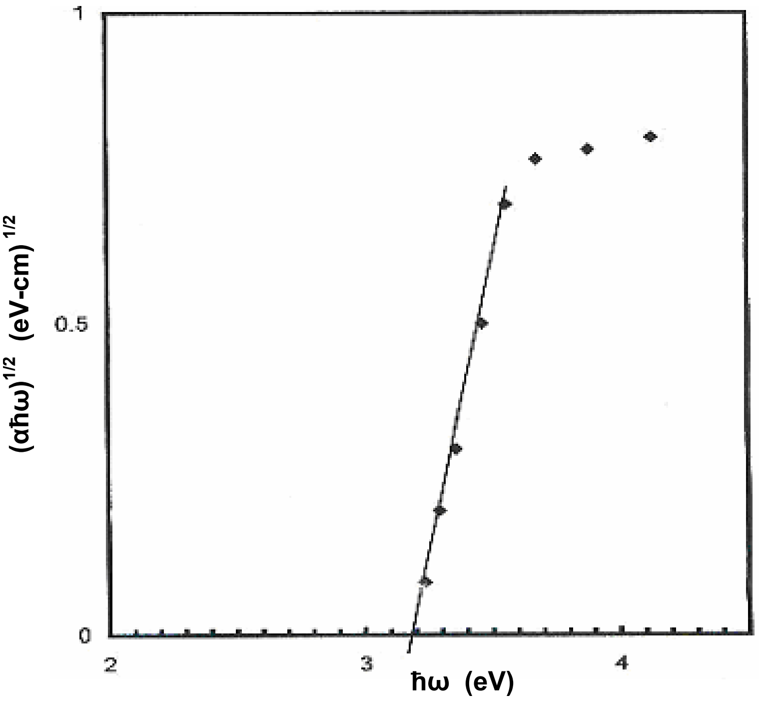

Fig. 7: Plot of $(\alpha \hbar \omega)^{1/2}$ versus photon energy $\hbar \omega$ to determine optical band gap of ZnO thin film (Thickness 3000Å, Evaporation Rate 5 Ás^{-1}).

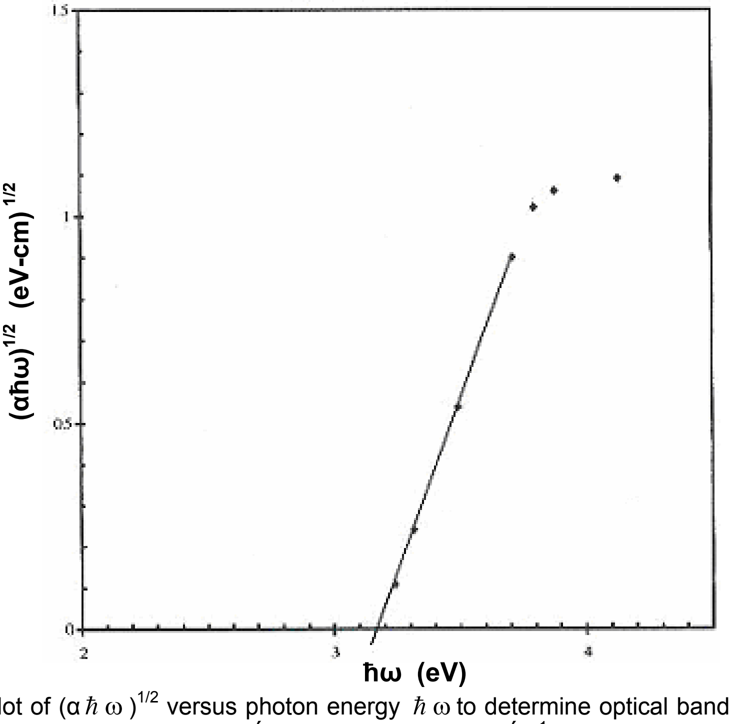

Fig. 8: Plot of $(\alpha \hbar \omega)^{1/2}$ versus photon energy $\hbar \omega$ to determine optical band gap of ZnO thin film (Thickness 3000Å, Evaporation Rate 10 Ás^{-1}).

References

Agashe, C., Kluth, O., Schope, G., Siekmann, H., Hupkes, J. and Rech, B. (**2003**) *Thin Solid Films*, **442**, 167.

Berger, L.I., Pamplin, B.P. (**1993**) "Properties of Semiconductors", In: R.C. Weast (Ed.), *Handbook of Chemistry and Physics*, 73rd ed., CRC Press, Boca Raton, FL, pp. 12-78 to 12- 85.

Chae, G.S. (**2001**) *Japanese Journal of Applied Physics*, Part 1, **40**, 1282.

Chaisitsak, S., Sugiyama, T., Yamada, A., Konagi, M. (**1999**) *Japanese Journal of Applied Physics*, Part 1, **38**, 4989.

Chen, Haiying; Qiu, Cheng Feng; Peng, Huajun; Xie, Zhilang; Wong, Man and Kwok, H.S. "Co-sputtered aluminum doped zinc oxide thin film as transparent anode for organic light-emitting diodes", Center for Display Research, Department of Electrical and Electronic Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China.

Fallah, H.R., Ghasemi, M., Hassanzadeh, A. and Steki, H. (**2006**) Applied Physics B **373**, 274-279.

France, Horiba (**2004**) "Spectroscopic ellipsometry study of ZnO thin films", Application Note.

Gorla, C.R., Emanetogla, N.W., Liang, S., Mayo, W.E., Lu, Y., Wrahack, M. and Shen, H. (**1999**) *Journal of Applied Physics*, **85**, 2595.

Jia, Chuan-Lei; Wang, Ke-Ming; Wang, Xue-Lin and Zhang, Xi-Jian (**2005**) "Formation of *c*-axis oriented ZnO optical waveguides by radio-frequency magnetron sputtering", *Optics Express*, **13**(13), 5093.

Kashani, H. (**1998**) "Structural, electrical and optical properties of zinc oxide produced by oxidation of zinc thin films", *Journal of Electronic Materials*, **27**(7), 876-882.

Kato, H., Sano, M., Miyamoto, K. and Yao, T. (**2002**) *Journal of Applied Physics*, **92**, 1960.

Manno, D., Micocci, G., Rella, R., Serra, A., Taurino, A. and Tepore, A. (**1997**) *Journal of Applied Physics*, **82**(1), 54.

Minami, T., Nanto, H., Takata, S. (**1985**) "Optical properties of aluminum doped zinc oxide thin films prepared by RF magnetron sputtering", *Japanese Journal of Applied Physics,* **24**(2), L605-L607.

Myoung, J.M., Yoon, W.H., Lee, D.H., Yun, I., Bae, S.H. and Lee, S.Y. (**2002**) *Japanese Journal of Applied Physics*, Part 1, **41**, 28.

Nakahara, K., Tanabe, T., Takasu, H., Fons, P., Iwata, K., Yamada, A., Matsubara, K., Hunger, R. and Niki, S. (**2001**) *Japanese Journal of Applied Physics*, Part 1, **40**, 250-254.

Ohgaki, Takeshi; Kawamura, Yuji; Kuroda, Takashi; Ohashi, Naoki; Adachi, Yutaka; Tsurumi, Takaaki; Minami, Fujio and Haneda, Hajime (**2003**) "Optical properties of heavily aluminum-doped zinc oxide thin films prepared by molecular beam epitaxy", *Key Engineering Materials,* **248**, 91-94, Sakai, K., Kakeno, T., Ikari, T., Shirakata, S., Sakemi, T., Awai, K. and Yamamoto, T. (**2006**) *J. Appl. Phys.*, **99**, 43508.

Sang, B., Dairiki, K., Yamada, A. and Konagi, M. (**1999**) *Japanese Journal of Applied Physics*, Part 1, **38**, 4983.

Srikant, V. and Clarke, D.R. (**1997**) "Anomalous behavior of the optical band gap of nanocrystalline zinc oxide thin films", *J. Mater. Res*., **12**(6), 1425.

Wang, Zhong Lin (**2004**) "Nanostructures of ZnO", *Materials Today*, **7**(6), 26-33.

Yoshino, Yukio; Ushimi, Yoshimitsu; Yamada, Hajime; Takeuchi, Masaki (**2003**) "Zinc oxide piezoelectric thin films for bulk acoustic wave resonators", Murata Manufacturing Co., Ltd., 2-26-10 Tenjin, Nagaoka-kyo, Kyoto, Japan.