



Highly luminescent ultra small Cu doped ZnO nanostructures grown by ultrasonicated sol–gel route



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ABSTRACT

Highly luminescent ultra small copper doped ZnO nanostructures were synthesized by ultrasonicated sol–gel route. Self assembled nanostructures were obtained on glass substrates using spin coater. The samples were characterized by XRD, AFM, Optical transmission, PL, FTIR and magnetic studies. Continuous ultrasonication helped in obtaining ultra small nanostructures. Observation of weak broadened peaks in X-ray diffractograms indicates the formation of nanocrystal of very small size ZnO. AFM micrographs show assembling of particles in linear direction resulting in formation of nanowires with increasing Cu concentration. The strong blue shift in the optical band gap of copper doped ZnO from 3.2 to 4.76 eV confirms the ultra small size of particles. Doping of copper not only improves the photoluminescence peak in UV region but also results in strong yellow, green and blue luminescence peaks. Undoped ZnO exhibits the diamagnetic behavior, however for higher doping concentration weak ferromagnetic phase observed. Strong green luminescence with weak ferromagnetic properties of ZnO nanowires makes them suitable for magnetic, optoelectronic and biological sensing devices.

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1. Introduction

Zinc oxide (direct band gap 3.37 eV) is one of the most important functional materials due to their special geometry and remarkable chemical-physical properties [1]. Luminescence property of ZnO nanoparticles are of great interest because of their strong dependence on the surface defects and size [2]. ZnO exhibits band edge emission in the near UV region and defect related emission in the visible region. High transmission in visible region and excellent conductivity make them promising candidate for transparent conducting electrodes and optoelectronic devices such as light emitting diodes, photo detectors, solar cells, spintronics, optical and magnetic sensors [3,4]. Photocatalytic activities are also reported in ZnO nanostructures with high green emission intensity [5]. Defects in ZnO nanostructures play crucial role in enhancing performance of optoelectronics devices. Recently lot of research is going on band gap engineering for the design of ZnO based short wavelength devices [6]. Improvement in various physical properties of ZnO is reported with doping of transition metals [7,8]. As the transition metals exhibit luminescence property it can also be used for modifying luminescence in ZnO nanostructures. Out of various transition

metals copper is one of the most promising dopant because of its comparable size with Zn ion, which can easily be substitute it. Cu behaves as acceptor in ZnO crystal make it good candidate for p type ZnO. Cu doped ZnO also attracts much interest due to potential application in spintronics [9]. It could modify luminescence in ZnO by creating localized impurity levels [10]. Synthesis of ZnO nanostructures has been reported using various deposition techniques like pulsed laser deposition, sputtering, MOCVD, electrodeposition, hydrothermal methods, sol gel and spin coating [11,12]. However for favoring self assembly of the nanostructures, sol gel proves to be a simple, low cost method known for high reproducibility, high controllability of composition and suitable for large scale production. In this paper we have reported the luminescence enhancement in Cu doped ZnO ultra small nanostructures grown by ultrasonic assisted low cost sol–gel route suitable for various optoelectronic applications.

2. Experimental detail

ZnO nanostructures were obtained by modified sol–gel chemical route. The Zinc acetate was (0.5 M) mixed with Ethanol and magnetically stirred for 60 min at room temperature. The sol, thus was kept in ultrasonic chamber at 40 kHz at 60 °C for another 2 h. For Cu doping Copper chloride was added in the ratio of 0.01 M (Sample A), 0.02 M (Sample B) and 0.03 M (Sample C). Mixture was magnetically

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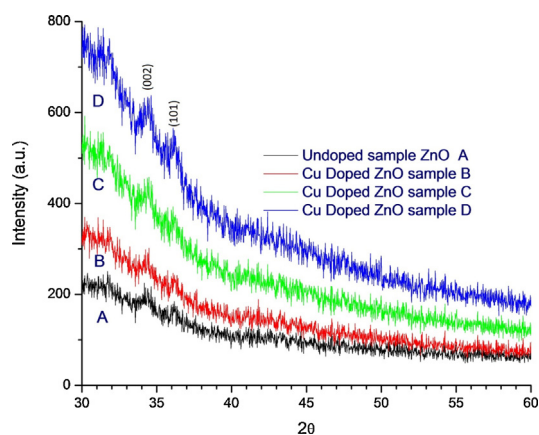


Fig. 1. X-ray diffractograms of undoped and Cu doped ZnO nanostructures.

stirred and heated to 60 °C for 2–3 h to obtain homogeneous solution. Gel was obtained by sonicating the sol for another 3 h at 60 °C followed by aging of 24 h. Particles were obtained by centrifuge the gel which was self assembled on glass substrates using spin coating. The samples were characterized by XRD (X-ray diffraction), AFM (atomic force microscopy), UV–vis transmission and PL (photoluminescence) FTIR (Fourier transform infra red spectroscopy) and magnetic studies. The transmission spectra were obtained using Perkin Elmer Lambda 25 in the range 200 to 800 nm. The PL spectra were obtained by Perkin Elmer LS55 with exciton at 200 nm of Xe lamp. The X-ray diffractograms were obtained in the 2θ range from 20 to 80° with Cu $K\alpha$ radiation of wavelength 1.546 Å using Bruker D8 Advanced XRD. AFM micrographs were obtained by using digital Instrumentation – Inc, USA. The FTIR spectra using Spectrum 2 and Magnetic studies were measured at room temperature using Lakeshore 7304 vibrating sample magnetometer (VSM).

3. Results and discussion

3.1. Structural studies

Fig. 1 shows the X-ray diffractograms of undoped and Cu doped ZnO nanostructures grown by ultrasonic sol–gel method. Small peaks of (1 0 0) and (0 0 2) are observed and identified as ZnO peaks [13]. Small intensity of the peaks indicates the formation of ultra small structures. Higher background at lower angles may be due to very thin layers grown on glass substrate similar results were also reported by others for very thin layers [14].

3.2. Surface studies

Atomic force micrographs for undoped and Cu doped ZnO nanostructures were shown in Fig. 2 [15]. Large numbers of islands are

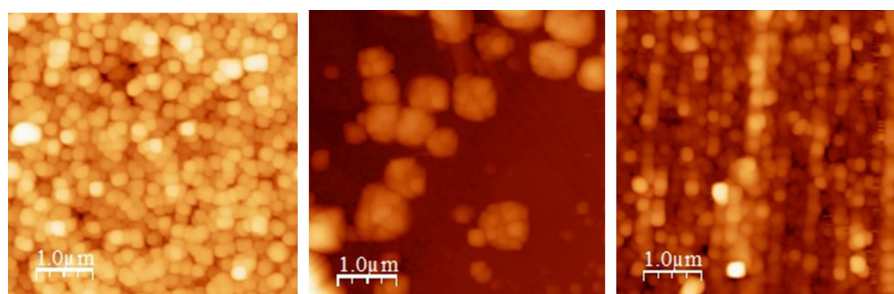


Fig. 2. AFM micrographs of (a) undoped ZnO, (b) & (c) Cu Doped ZnO (samples A and C).

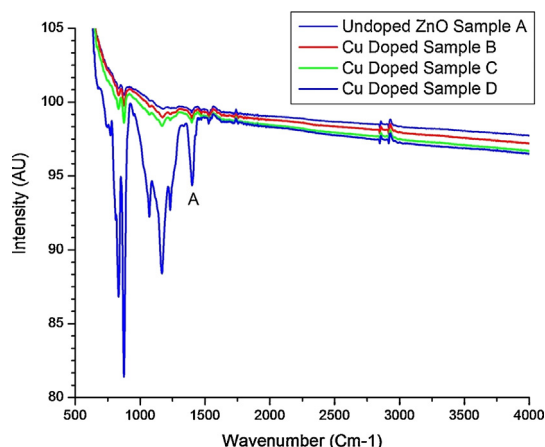


Fig. 3. FTIR spectra of undoped ZnO and Cu doped ZnO nanostructures.

observed in undoped ZnO. Observable feature of these structures is grin like. On doping with copper these islands grew as more and more particles bound together. Further addition of Cu results in alignment of all islands preferably with vertical orientation.

3.3. Spectroscopic studies

3.3.1. FTIR

As more atoms/molecules are arranged on the surface of nanoparticles, the surface chemistry of these molecules is of immense interest in order to establish the presence or absence of various vibration modes in ZnO nanoparticles at room temperature, and to study the effect of doping, FTIR spectra for ZnO and Cu doped ZnO have been obtained shown in Fig. 3.

1573 cm^{-1} and 1403 cm^{-1} are related to symmetrical and asymmetrical stretching modes of carboxylic group of acetate coordinated to the surface of ZnO might be due to starting material zinc acetate as source [16,17]. The ZnO nanocrystals terminated with acetate group which confirm long term colloidal stability. Stretch of band appearing at 666 cm^{-1} confirms the formation of rod shape ZnO particle. 780 cm^{-1} and 1050 cm^{-1} shows specific fingerprint of ZnO already reported by others (Elbey and Samer). The peaks around 2830 cm^{-1} and 2925 cm^{-1} are due to C–H bond bending and bond stretching, respectively. Observation of such bands suggest the presence of absorbed species on the surface (surface modification) of the nanocrystals. Absence of absorption peak of –OH group of H_2O , indicates non existence of water absorbed on the surface of nanocrystalline powders.

3.3.2. Optical studies

Fig. 4 shows the Optical transmission spectra for undoped and doped ZnO particles. Constant absorption coefficient for ZnO

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