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Tailoring the photoluminescent property of ZnO/Ag nanocomposite thin films based on a thermal treatment

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ABSTRACT

It is a recent hot topic of widespread interest to control the optical properties of metal oxide by noble metal nanoparticles. In this work, ZnO/Ag nanocomposite thin films were prepared by one-step sol-gel method. The crystal phase, morphology of the samples was analyzed by X-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. The presence of silver nanoparticles was confirmed by transmission electron microscopy (TEM). The distribution and chemical valence state of Ag in the samples was analyzed by elemental mapping images and X-ray photoelectron spectroscopy (XPS), respectively. The optical properties of the samples were studied by UV–visible spectroscopy and photoluminescence. It is found that the thermal treatment temperature has an important effect on the chemical state of silver in ZnO, and the change of chemical state of silver in turn affects the ultraviolet (UV) emission of ZnO. Under an appropriate heating temperature, the doped silver in the ZnO thin film can form silver nanoparticles, and the photoemission transfer of electrons from the silver nanoparticles to ZnO can effectively enhance the UV emission of ZnO thin film. The results show that under the experimental conditions applied in this study, 250°C is an optimum heating temperature, since the ZnO/Ag composite film heated at 250 °C has the strongest UV emission performance.

1. Introduction

In the last decade, zinc oxide (ZnO), as a multifunctional semiconductor material, has inspired a lot of research enthusiasm in the world [1–6]. And its luminous behavior seems to attract more attention [7–11], since ZnO has a wide direct bandgap of ∼ 3.37 eV at room temperature with a large excitonic binding energy of ∼ 60 meV which allows ZnO nanomaterials to have high exciton emission efficiency at room temperature even at higher temperatures. Moreover, compared with GaN, the growth temperature for high-quality ZnO epitaxial film is lower, leading to lower production costs. These advantages make ZnO nanomaterials an ideal candidate for the development of large-scale short-wavelength light-emitting devices. In order to get higher exciton emission efficiency from ZnO, many strategies have been adopted, such as optimizing deposition parameters and annealing conditions for ZnO materials [12–14], doping [15,16], surface modification [17], etc. In recent years, it has been found that the noble metal nanoparticles can greatly enhance the UV emission performance of ZnO [1,18–20]. For example, Lai et al. [18] prepared Ag-capped ZnO thin films by sputtering and thermal evaporation and studied the influence of Ag-capping layer on the UV emission of ZnO thin films. They found that the Ag-capping layer could greatly improve UV emission of ZnO thin films and attributed this to the resonant coupling of the spontaneous emission in ZnO into the surface plasmons of Ag to enhance the emission efficiency. Lawrie et al. [1] found that the ZnO band-edge emission enhancement disappeared when a 10 nm MgO spacer layer separated Ag (Au) and ZnO thin films, which provided an evidence for hot electron transfer across the metal-ZnO interface as the PL enhancement mechanism. Ren et al. [20] used Ni to coat ZnO nanowire and found that the UV emission of ZnO was increased; the enhancement was attributed to energy transfer between near-band-edge (NBE) emission in ZnO and surface plasmons in the Ni coating layer. Liang et al. [21] fabricated ZnO-Ag composite thin films with various degrees of Ag addition by varying the Ag sputtering power level from 3 to 13 W; they found that the UV emission band dominated the 9 W Ag co-sputtered thin film and the enhancement of the UV emission was attributed to effective Ag doping which reduced the density of zinc and oxygen vacancies in ZnO lattices. Recently, Pal et al. [22] prepared ZnO nanorods on Ag thin film by hydrothermal method. They deemed that the enhanced UV emission from ZnO/Ag composites should result from two factors: (1) the

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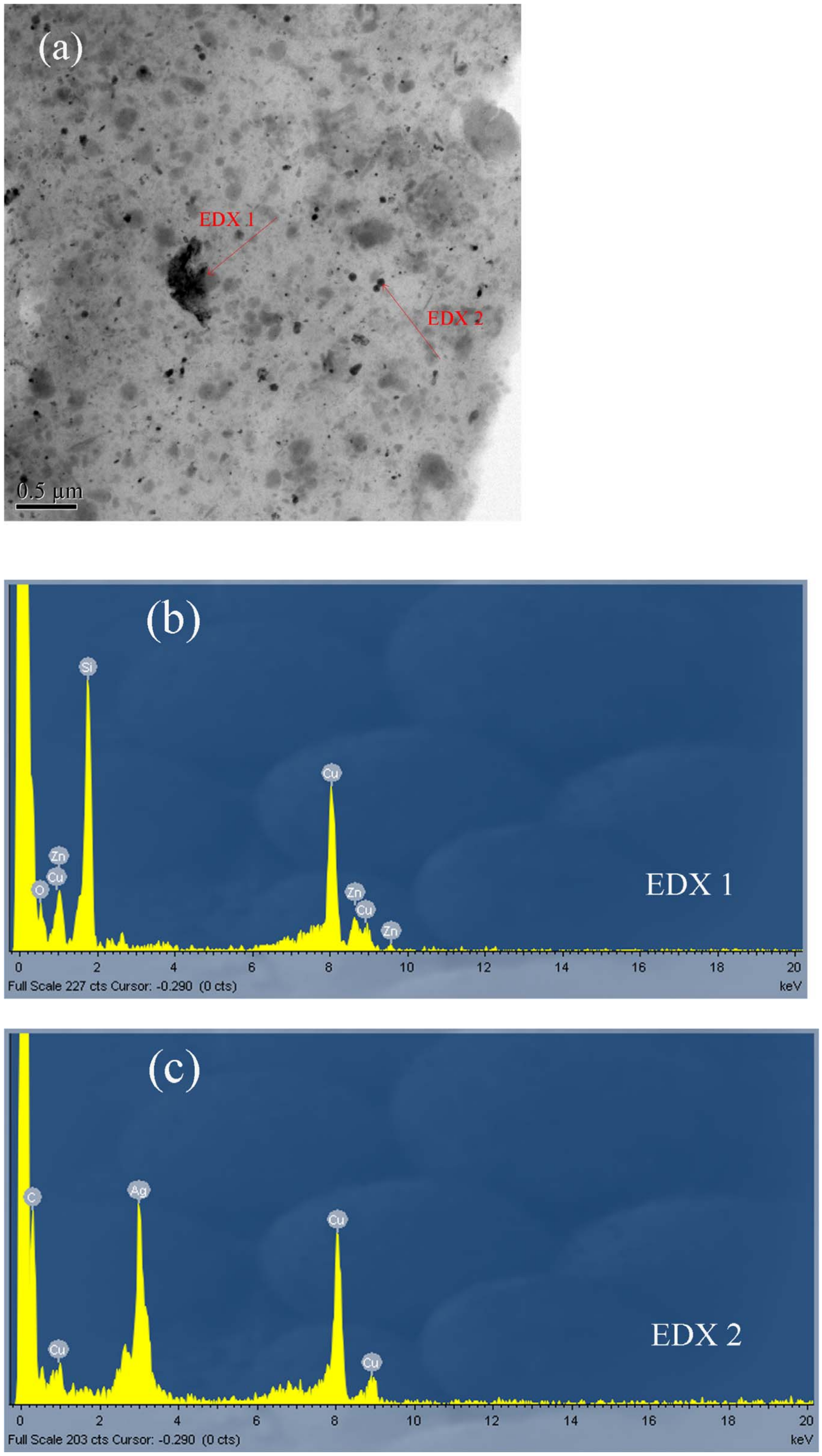

Fig. 1. TEM image (a) and EDX spectra (b, c) of the sample heated at 200°C.

resonant SP-exciton coupling occurs at the ZnO/Ag interface; (2) the transfer of charge carriers between ZnO and Ag due to their direct contact increases electron density in the conduction band of ZnO and leads to the increase of electron-hole recombination rate. Although the understanding of the UV emission enhancement mechanism in the ZnO/Ag system is still inconsistent, many experiments have proved that Ag nanoparticles can enhance the UV emission performance of ZnO.

In the Ag/metal oxide system, the chemical state of Ag has a great influence on the optical properties of metal oxides, while the chemical state of Ag is in turn strongly dependent on the thermal treatment conditions [22–24]. For example, Viana et al. [24] deposited Ag/TiO_2 nanocomposite thin films by sol-gel method and annealed them; they found that the mechanism of migration, segregation and growth of Ag nanoparticles observed depended on the thermal treatment temperature and the cooling rate. As for the ZnO/Ag nanocomposites, it is necessary to deeply investigate the influence of thermal treatment on the