

Enhancement of ultraviolet lasing from Ag-coated highly disordered ZnO films by surface-plasmon resonance

A. P. Abiyasa, S. F. Yu,^{a)} S. P. Lau, Eunice S. P. Leong, and H. Y. Yang

School of Electrical and Electronic Engineering, Nanyang Technological University, Block S2, Nanyang Avenue, Singapore 639798, Singapore

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Large improvement in random lasing action at ultraviolet wavelength has been achieved from highly disordered ZnO films with Ag coating. The lasing threshold can be reduced by two times and slope efficiency can be increased by 5.5 times. The improvement is due to the presence of Ag coating, which enhances the surface coupling of lasing emission from the ZnO films by surface-plasmon resonance and reduces the scattering loss experienced by the random cavity modes. Furthermore, the enhancement of lasing efficiency is dependent on the Ag coating's surface roughness, which can be controlled through the surface morphology of ZnO films. © 2007 American Institute of Physics. [DOI: 10.1063/1.2746940]

ZnO has attracted considerable attention to realize high internal quantum efficiency ultraviolet (UV) lasers due to its wide band gap and high excitonic binding energy at room temperature.^{1,2} However, it is still necessary to improve the external conversion efficiency of the lasers in order to achieve better performance. Recently, enhancement of spontaneous emission efficiency has been obtained by surface-plasmon mediated surface emission from rough metal surface.^{3,4} However, this has yet been demonstrated in ZnO for the improvement of UV lasing emission. In this letter, we studied the possibility to enhance UV lasing efficiency from highly disordered ZnO films by surface-plasmon resonance (SPR). It was found that the presence of rough Ag coating on ZnO films supports SPR and random lasing action simultaneously so that the corresponding lasing performance can be improved. On the other hand, reduction of lasing intensity was observed from the Ag-coated ZnO films with relatively smooth surface (i.e., suppression of SPR coupling). Hence, we have verified that SPR can be used to facilitate UV lasing from ZnO lasers.

Two sets of ZnO films, which have thicknesses of ~ 200 and ~ 120 nm, were deposited onto quartz substrates by filtered cathodic vacuum arc technique at room temperature. The films were annealed at ~ 900 °C for ~ 2 h in open air to form highly disordered ZnO films to sustain random lasing action.⁵ As the two sets of ZnO films have different thicknesses, the same annealing process will produce ZnO grains with different sizes and densities. It is expected that thinner ZnO film can produce smaller and more closely packed grains (i.e., rougher surface morphology) than that of the thicker one. The ZnO films were then cut into few ~ 1 cm² samples for the deposition of Ag by Ar⁺ ion-beam sputtering. The ion-beam energy and current of the Kaufmann-type ion-beam source were set to ~ 1 keV and 60 mA, respectively, during the deposition. The deposition rate of Ag was found to be ~ 2 nm/min at a chamber pressure of $\sim 3 \times 10^{-4}$ Torr. A transmission electron microscope (TEM) cross-section image of an annealed ZnO (~ 200 nm) film coated with ~ 40 nm thick Ag is shown in Fig. 1(a). It is noted that the

slow deposition rate of Ag coating preserved the surface profile of ZnO grains. The atomic force microscope (AFM) images before and after the deposition of Ag coatings have shown similar surface morphology. This indicated that the surface morphology of Ag coating follows the surface profile of ZnO films. Hence, by controlling the size and density of ZnO grains, different densities of surface roughness of Ag coating can be obtained.

Figure 1(b) shows the schematic of the optical measurement setup used to analyze the emission characteristics of the ZnO samples. A frequency-tripled neodymium doped yttrium aluminum garnet pulsed laser (355 nm, 10 Hz) with spot size of ~ 1 mm in diameter was used to excite the samples at room temperature. Large beam diameter (i.e., low optical density) is used to avoid laser ablation of the Ag coatings. Radiation emitted from the surface of the samples was then collected through a pinhole of diameter of ~ 250 μ m at different emission angles θ by a monochromator-detector setup. Hence, the spectra of the surface radiation observed from the samples were studied spatially using the setup. To prevent oxidation of the Ag coating, the samples were kept inside vacuum prior to the measurement.

Figure 2(a) plots the spontaneous emission intensities versus θ of the ~ 200 nm thicknesses ZnO films without and with different thicknesses of Ag coating. The samples were optically pumped at ~ 0.1 MW/cm² (i.e., below lasing threshold). Only half of the emission profile is plotted since

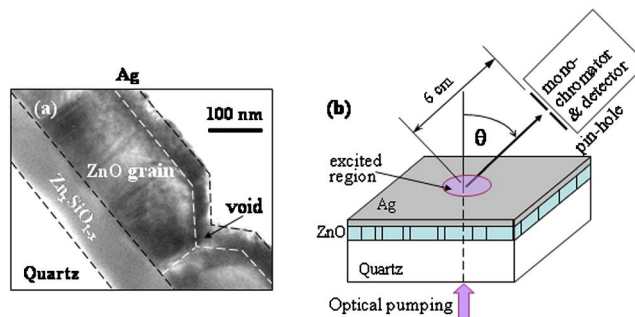


FIG. 1. (Color online) (a) TEM image of a Ag-coated (~ 40 nm) ZnO (~ 200 nm) film deposited on quartz substrate. (b) Schematic diagram of the optical measurement setup.

^{a)} Author to whom correspondence should be addressed; electronic mail: esfyu@nut.edu.sg

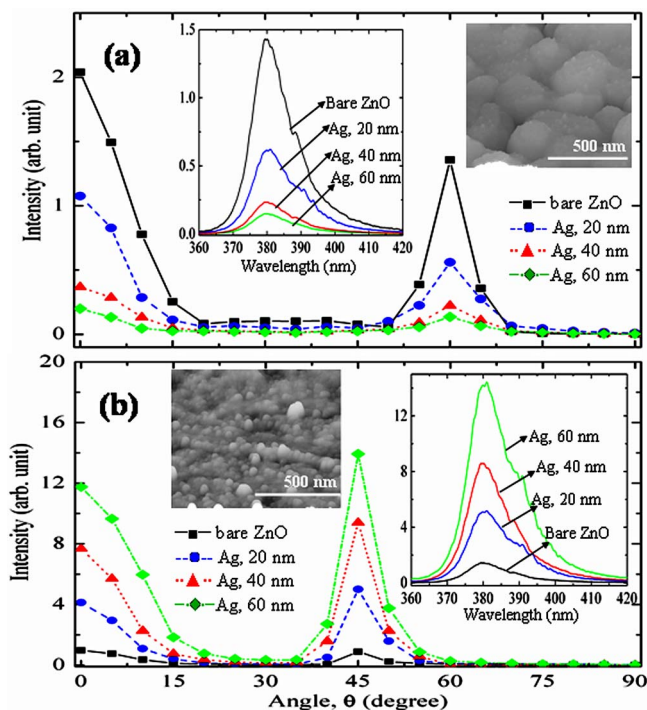


FIG. 2. (Color online) (a) Spontaneous emission intensity of ~ 200 nm thickness Ag-coated ZnO films vs emission angle θ . Inset: (left) spontaneous emission spectra measured at $\theta \sim 60^\circ$ and (right) AFM image of an ~ 20 nm thick Ag layer deposited on an ~ 200 nm thick ZnO film. (b) Spontaneous emission intensity of ~ 120 nm thickness Ag-coated ZnO films vs emission angle θ . Inset: (right) spontaneous emission spectra measured at $\theta \sim 45^\circ$ and (left) AFM image of an ~ 20 nm thick Ag layer deposited on an ~ 120 nm thick ZnO film. Emission characteristics of bare ZnO films are also included for comparison.

the emission profile is symmetric about the normal direction. It is observed that two distinct spontaneous emission peaks emerged at $\theta=0^\circ$ and $\sim 60^\circ$. The emission peak emitted at $\theta=0^\circ$ resembles the typical dipole emission of direct band gap semiconductor materials. The peak at $\theta \sim 60^\circ$ is due to diffraction of the spontaneous emission by the random distribution of ZnO grains. The inset of the figure plots the corresponding spontaneous emission spectra at $\theta \sim 60^\circ$. The peak emission is centered at ~ 380 nm which corresponds to free-exciton recombination of ZnO at room temperature. It is noted that the emission spectra at $\theta \sim 0^\circ$ is similar to that at $\theta \sim 60^\circ$. On the other hand, the intensity of surface spontaneous emission reduces with the increase of Ag thickness for all emission direction. For Ag coating thicker than 60 nm, most of the emission was either reflected or absorbed at the Ag/ZnO interface.

Figure 2(b) plots the spontaneous emission intensities versus θ the ~ 120 nm thickness ZnO films without and with different thicknesses of Ag coating. It is observed that two distinct spontaneous emission emerged at $\theta=0^\circ$ and $\sim 45^\circ$. Furthermore, the spontaneous emission intensity increases with the thickness of Ag and the enhancement of emission intensity can be more than ten times for the thick Ag coating. This is because thick Ag coating adjusts the SPR energy closer to that of the ZnO emission and hence increases the Purcell enhancement factor.⁶ The inset of the figure plots the corresponding spontaneous emission spectra at $\theta \sim 45^\circ$. It is found that the enhancement of surface emission is dependent on the surface roughness of Ag which can be controlled by the ZnO surface morphology. AFM images of the ~ 20 nm

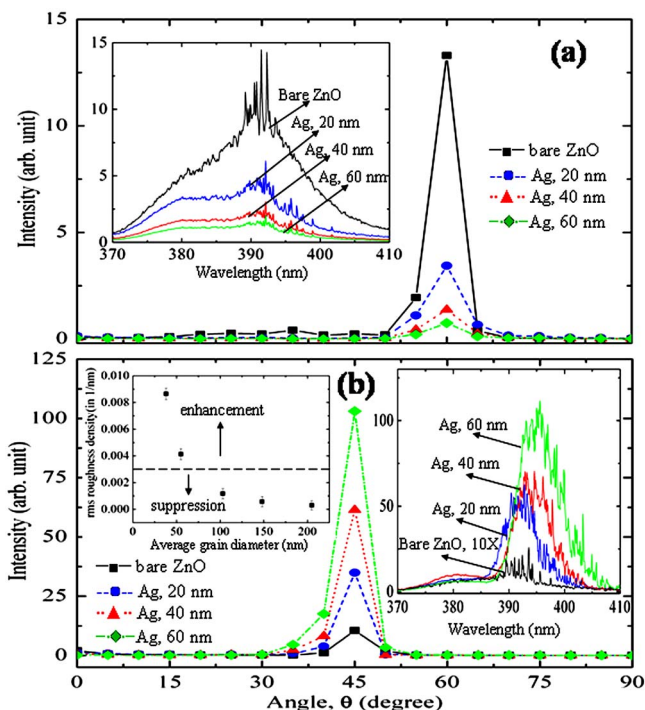


FIG. 3. (Color online) (a) Lasing emission intensity of ~ 200 nm thickness Ag-coated ZnO films vs angle θ . Inset: lasing emission spectra measured at $\theta \sim 60^\circ$. (b) Lasing emission intensity of ~ 120 nm thickness Ag-coated ZnO films vs angle θ . Inset: (right) lasing emission spectra measured at $\theta \sim 45^\circ$ and (left) rms roughness density of Ag-coated surface vs average ZnO grain size. Lasing characteristics of bare ZnO films are also included for comparison.

thick Ag surface coated on the ZnO films have shown that only the highly dense rough Ag surface on ZnO films with average ZnO grain size of ~ 50 nm [i.e., inset of Fig. 2(b)] will exhibit enhancement of surface emission. However, relatively less dense rough Ag surface on ZnO films with average ZnO grain size of ~ 150 nm [i.e., inset of Fig. 2(a)] suppresses surface emission. In fact, these observations are consistent with the recent studies on the use of rough metal surface to enhance surface emission.^{4,6}

We have shown that the formation of highly disordered ZnO films can provide a template to form rough Ag coating for the excitation of SPR. The presence of ZnO grains and voids can also sustain random lasing action inside the highly disordered ZnO films.⁵ Hence, we further studied the random lasing characteristics of the Ag-coated ZnO films. Figure 3(a) plots the lasing intensities versus θ of the ~ 200 nm thickness ZnO films without and with different thicknesses of Ag coating. The samples were optically pumped at ~ 0.35 MW/cm² (above threshold). It is noted that lasing emission was only detected at $\theta \sim 60^\circ$. This value of θ is similar to the case of surface spontaneous emission given in Fig. 2(a). This verified that the diffraction of spontaneous emission is due to the presence of ZnO grains and voids. However, the presence of Ag coating suppresses the lasing intensities, and no lasing emission was detected for Ag coating thicker than ~ 60 nm.

Figure 3(b) plots the lasing intensities versus θ of the ~ 120 nm thickness ZnO films without and with different thicknesses of Ag coating. Random lasing action is only recorded at $\theta \sim 45^\circ$ [i.e., this value is similar to that measured from the spontaneous emission, see Fig. 2(b)], and the presence of Ag coating enhances lasing intensities especially for

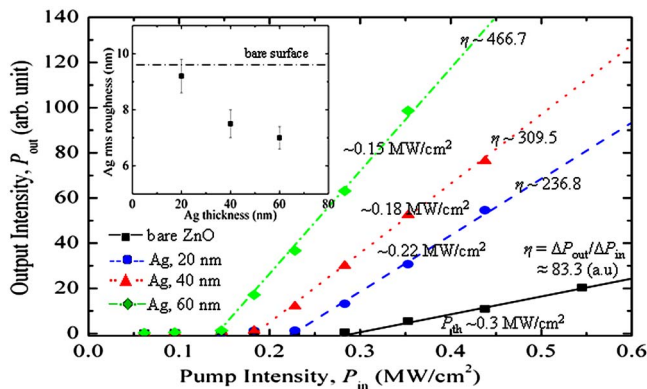


FIG. 4. (Color online) Light-light characteristics of ~ 120 nm thickness Ag-coated ZnO films. Inset: the surface rms roughness of ~ 120 nm thickness Ag-coated ZnO films vs different thicknesses of Ag coating. Lasing characteristics of bare ZnO films are also included for comparison.

thick Ag coating. This is because thick Ag coating increases the coupling of lasing emission from ZnO random cavities into surface emission via SPR. To determine the influence of surface roughness density (i.e., defined as rms roughness per grain area) on the enhancement of the UV lasing emission, the lasing experiment was repeated for Ag-coated ZnO samples with different ZnO grain sizes (i.e., by the fabrication of Ag-coated ZnO films with different thicknesses). The inset of Fig. 3(b) plots the corresponding rms roughness density of Ag (with thickness of ~ 20 nm) versus ZnO grain diameter. It is noted that there is a minimum value of rms roughness density that can sustain SPR for the enhancement of lasing emission from the rough Ag coating.

Figure 4 shows the light-light curves of the ~ 120 nm thick ZnO samples without and with Ag coating of different thicknesses. It is observed that the lasing threshold (slope efficiency) of sample can be reduced (increased) by more than 2 (5.5) times with the presence of thick Ag coating. The expression of lasing threshold P_{th} and slope efficiency η of the samples can be approximated by $P_{th} \propto \alpha_{scatt} + \alpha_{SPR} + L^{-1} \ln(1/R) + \alpha_{abs}$ and $\eta \sim (\alpha_{SPR} + L^{-1} \ln(1/R)) / (\alpha_{scatt} + \alpha_{SPR} + L^{-1} \ln(1/R))$, respectively, where α_{scatt} is the scattering loss experienced by the random cavity modes, α_{SPR} is the emission loss related to SPR, and α_{abs} is the internal absorption loss of ZnO. L is the length and R is the effective reflectivity of the random cavities. It is noted that the values of α_{abs} , L , and R will not be changed by the presence of Ag coating as they are mainly dependent on the

ZnO random cavities. As α_{SPR} is defined as the radiation loss related to the surface emission via SPR, the value of α_{SPR} should increase with the thickness of Ag. In order to satisfy the reduction (increase) of P_{th} (η) with the increase of Ag thickness, as observed in Fig. 4, the value of α_{scatt} should reduce with the increase of Ag thickness. As α_{scatt} is defined as the radiation loss not coupled to surface plasmons, the presence of Ag coating will reduce the value of α_{scatt} (i.e., improve the optical confinement of the random cavity modes). In addition, if the surface roughness of Ag coating can be reduced with the increase of thickness of Ag coatings, the value of α_{scatt} should also be reduced. The inset of Fig. 4 plots the rms surface roughness of the Ag deposited on the ~ 120 nm thick ZnO sample versus the thickness of Ag coating. It is noted that the surface rms roughness reduces with the increase of the Ag thickness as Ag fills up the voids. This indicated that the presence of Ag coating reduces the surface roughness of air/Ag interface so that the value of α_{scatt} should reduce with the increase of Ag thickness. However, the surface roughness is rough enough to sustain SPR.

In conclusion, we have verified that UV random lasing action obtained from highly disordered ZnO films can be improved significantly by SPR. This can be realized by depositing Ag coating on the surface of ZnO films with average grain diameter of ~ 50 nm or less. As a result, both SPR and random lasing action can be excited simultaneously inside the Ag-coated ZnO films. It is noted that the Ag thickness of ~ 60 nm is sufficient to improve the lasing efficiency of the ZnO films by more than a double. Furthermore, the improvement of lasing performance can be interpreted as the increase of spontaneous emission via SPR (i.e., increase α_{SPR}) as well as the improvement of optical confinement (i.e., reduce α_{scatt}) of the random cavity modes.

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