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Synthesis and optical characterization of pure and nickel doped gallium nitride nanocrystals

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ABSTRACT

In the present Letter, $Ga_{1-x}Ni_xN$ ($0 \le x \le 0.1$) nanocrystals have been synthesized by facile solvothermal method. Crystallographic characterization of synthesized materials has been done, using powder X-ray diffraction technique, reveal the formation of wurtzite (hexagonal) structured GaN nanocrystals. Electron microscopic studies have been carried for the detailed topographical and morphological analysis of synthesized nanomaterials. Spectroscopic studies like energy dispersive X-ray spectroscopy (EDS), UV–visible absorption, X-ray photo electron spectroscopy (XPS), steady state energy resolved and time resolved photoluminescence (PL) spectroscopy have been opted for the investigation of quantitative, qualitative and optical behavior of synthesized nanomaterials.

1. Introduction

Semiconductor nitrides such as, gallium nitride (GaN), aluminum nitride (AlN) and indium nitride (InN) are very promising materials for their potential use in opto-electronic devices as well as high-power and high-temperature electronic devices [1,2]. These materials and their ternary and quaternary alloys cover a wide energy band gap range of 1.9–6.3 eV, suitable for band-to-band light generation with colors ranging from red to ultra-violet wavelengths. Nitrides are particularly suitable for applications in surface acoustic wave devices, UV detectors, Bragg reflectors, waveguides, and UV and visible light emitting diodes (LEDs) [3]. During these days, GaN nanostructures seem to be accepted as the most important materials after silicon for opto-electronic and electronic applications due to their size tunable characteristic properties caused by quantum confinement and augmentation of surface to volume ratio. The optical properties of GaN are of great interest due to its potential applications for the fabrication of laser diodes [4,5], color tunable light emitting diodes [6], high power electronic devices [7] flat panel displays, optical data storage devices, photodetectors [8] and smart nano sensors [9] due to its direct wide band gap.

There are various methods to grow one dimensional GaN nanostructures such as laser induced growth [10], template assisted synthesis [11], catalyst assisted synthesis [12], oxide assisted method [13], the reaction of gallium and ammonia gas [14,15], pyrolysis [16], vapor-phase epitaxy [17] and chemical vapor deposition method [18]. However, all these methods require critical reaction conditions and this limits the application field.

In the present Letter, a low cost and a low temperature eco-friendly solvothermal synthesis strategy has been opted to synthesize pure and Ni doped GaN nanocrystals. Crystallographic, morphological and optical characteristics of synthesized nanomaterials have been thoroughly studied to explore their potential for next era smart opto-electronic industrial applications. To the best of our knowledge, this Letter is the first report mentioning the optical properties of $Ga_{1-x}Ni_xN$ ($0 \le x \le 0.1$) nanostructures.

2. Experimental

For the synthesis of $Ga_{1-x}Ni_xN$ ($0 \le x \le 0.1$) nanostructures, analytical grade chemicals: Gallium (II) chloride (Ga_2Cl_4), Nickel (II) nitrate hexahydrate $[Ni(NO_3)_2]{\cdot}6H_2O$, hexamethyldisilazane ($C_6H_{19}NSi_2$), and toluene were procured from Sigma–Aldrich. All these chemicals were used without any further purification for the synthesis of pure and doped GaN nanostructures by solvothermal method.

In a typical synthesis process, $Ga_{1-x}Ni_xN$ nanocrystals formation involves the solvothermal decomposition of Ga_2Cl_4 and $[Ni(NO_3)_2]{\cdot}6H_2O$ in the presence of hexamethyldisilazane (HMDS) and cetyltrimethylammoniumbromide (CTAB). To synthesize pure and Ni doped GaN nanocrystals stock solution of Ga_2Cl_4 was prepared by dissolving 1 g of Ga_2Cl_4 in 144.7 ml of toluene, whereas,

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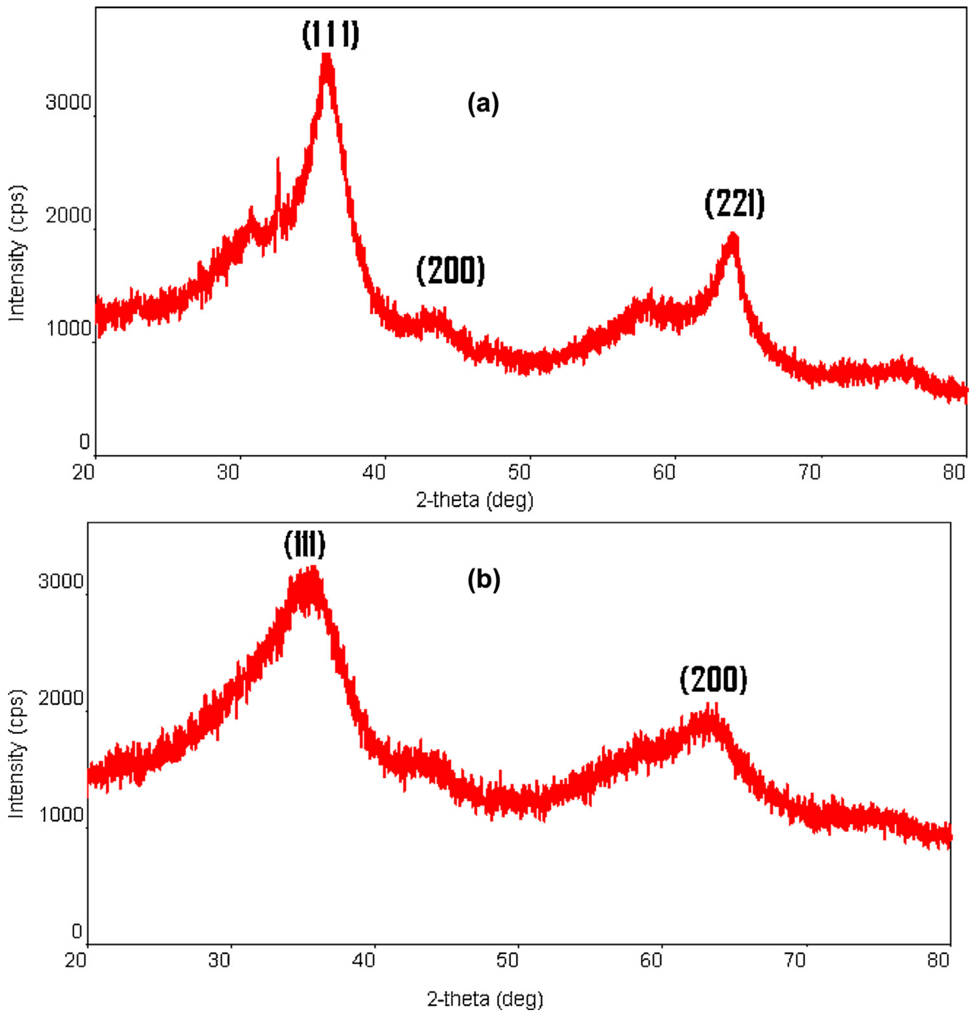

Figure 1. (a) XRD pattern of pure and (b) 1% Ni doped GaN nanocrystals.

nickel precursor 0.05 M solution was prepared by dissolving 0.1455 g of $[Ni(NO_3)_2]{\cdot}6H_2O$ in 10 ml ethanol. For the preparation of pure GaN nanocrystals, mixture of 43.5 ml of Ga_2Cl_4 and 22 ml of HMDS solutions, along with 0.45 g of CTAB, was taken in a Teflon-lined autoclave. The resulting mixture solution was treated at 240 °C in the sealed autoclave for 3 h and then was allowed to cool down for 10 h. The resultant product was washed several times to remove unwanted impurities and was dried at 70 °C for 12 h. The same process was followed for the preparation of 10%, 1% and 0.1% Ni doped GaN nanostructures.

3. Characterization

Powder X-ray diffractometer (Rigaku Miniflex-600 XRD) was used to record the X-ray diffraction patterns of the synthesized materials for crystallographic characterization. Diffraction patterns were recorded for all the samples in the 2θ range 20–80 °C by keeping the step size 0.02° at the scan speed 4 deg/min, at the generator tension of 40 kV and generator current 15 mA. Field emission scanning electron microscope (SUPRA 55 OXFORD INSTRUMENTS), equipped with energy dispersive X-ray spectrometer (EDS), was used for the topographical as well as qualitative and quantitative analysis. The synthesized powder samples were loaded on microscopic stub with the help of double adhesive tape followed by gold coating for FESEM analysis.

TEM (Hitachi make) and HRTEM (TECNAI-G2 2005) were used for morphological analysis of synthesized nanomaterials. Sample preparation for TEM/HRTEM analysis involves the dispersion of nanopowders in ethanol/water followed by ultrasonication, and then a drop of sample solution was loaded on a carbon coated copper grid and 2% amyl acetate coated Ni grid for TEM and HRTEM studies, respectively.

Optical characterization of synthesized nanomaterials was done by UV–visible absorption, Fourier transform infra-red spectroscopic studies, X-ray photo electron spectroscopy and steady state and time resolved PL spectroscopy. Hitachi U-2900 UV-vis. absorption spectrophotometer was used to record absorption spectra of nanomaterials dissolved in alcoholic (ethanol) media. X-ray photoelectron spectroscopy (XPS) was performed using kratos axis ultra DLD. A room temperature steady state photoluminescence spectrum was recorded using SHIMADZU Spectrofluorophotometer (RF-5301 PC). The $Ga_{1-x}Ni_xN$ nanocrystals were excited at excitation wavelength of 325 nm using xenon lamp as the excitation source. Time resolved fluorescence spectroscopy of these nanomaterials was performed using Edinburgh FL 920 Fluorescence life time spectrometer.

4. Results and discussion

Solvothermal synthesis method, opted in the present investigation, is an eco-friendly facile bottom up synthesis technique, which gives good yield of nearly monodisperse intrinsic and extrinsic GaN nanostructures. Figure 1a shows XRD pattern recorded for pure GaN nanopowder synthesized by solvothermal method