MAGNETIC AND OPTICAL PROPERTIES OF MN DOPED GAN

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ABSRACT

Mn:doped GaN films, a dilute magnetic semiconductor material, are grown on (0001) sapphire substrates by metal organic vapor deposition. Optical properties are investigated by transmission measurements and two absorption bands are found to be dominating the transmission spectra. The first absorption band was at Ev+1.5 eV and was attributed to the formation of a Mn related deep level energy band above the valence band of GaN. The second band extended from 2.0 eV to the bandedge of GaN. Absorption at these bands scaled with thickness and composition of the films. Co-doping of these films by n-type (Si) and p-type dopants (Mg) also greatly enhanced or reduced the absorption at these bands, indicating a change in the Fermi level in the crystal. The magnitude of the saturation magnetization was also a function of the Fermi level, which was probed by transmission measurements. Therefore, a correlation between optical properties and saturation magnetization has been established.

1. INTRODUCTION

GaMnN has attracted much interest recently due to its potential as a dilute magnetic semiconductor (DMS) and a building block for spintronic devices (Dietl et al., 2000). Ferromagnetic GaMnN crystals exhibiting a range of Curie temperatures up to and above room temperature have been synthesized by several different growth techniques (Theodoropoulou et al., 2001; Thaler et al., 2002; Kim et al., 2003; Sonoda et al. 2002; Song et al., 2005; M. J. Reed et al. 2005; Arkun et al., 2004; M. L. Reed et al. 2001). Optical absorption studies on GaMnN films indicate that Mn forms a deep acceptor band with an optical transition at 1.5 eV and a broad absorption band that commences at 2.0 eV (Korotkov et al. 2001). Photoluminescence spectra of the same samples show infrared emission at 1.27 eV which further supports the position of the Mn band identified by optical absorption measurements. Others observed that introducing Mn into GaN led to the appearance of a very strong absorption band starting at 1.9eV and attributed this band to a transition from Mn acceptor states to the conduction band (Polyakov et al., 2002). The absence of the absorption band with a threshold of ~1.5eV was attributed to the Fermi level being above the Mn band. Additional reports on the optical absorption measurements of n-type GaN:Mn and GaN:Mn,Mg observed an absorption band with a threshold of 2.0eV tailing to the bandedge of GaN (Wolos et al., 2004). Wolos et al. also observed an additional absorption peak in the infrared region with two sharp peaks at ~1.41eV and 1.48eV in the case of GaN:Mn,Mg. These peaks were attributed to an internal transition of the Mn atom followed by thermal ionization to the GaN valence band. Although many reports on the optical properties are given in the literature, little information is available regarding the correlation between ferromagnetism, the effect of co-doping, and the optical properties of GaMnN films.

In this paper, optical transmission measurements on undoped, silicon doped and magnesium doped GaMnN samples will be presented and correlations between ferromagnetism and optical properties will be discussed.

2. EXPERIMENTAL

Growth of GaMnN samples was performed on (0001) sapphire substrates by metal organic chemical vapor deposition (MOCVD). Details of the growth reactor and growth conditions are reported elsewhere (M. J. Reed et al., 2005; Arkun et al., 2004). Optical transmission measurements were performed using a Varian Cary 5E UV-VIS-IR dual beam spectrometer with a spectral range between 175nm and 3.3μm. All measurements were performed at room at temperature. Emission from a tungsten halogen light source was used in line with a spectrophotometer to selectively scan wavelengths between 200nm and 900nm. Ultraviolet and visible light was detected by a Hamamatsu photomultiplier tube (PMT), whereas a lead sulfide detector was used for detection of infrared light.

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3. RESULTS AND DISCUSSION

Optical transmission measurements were performed on 1.4µm thick $Ga_{1-x}Mn_xN$ samples at 300K where (x=0.003-0.016). The transmission spectrum is plotted as a function of photon energy in Figure 1.

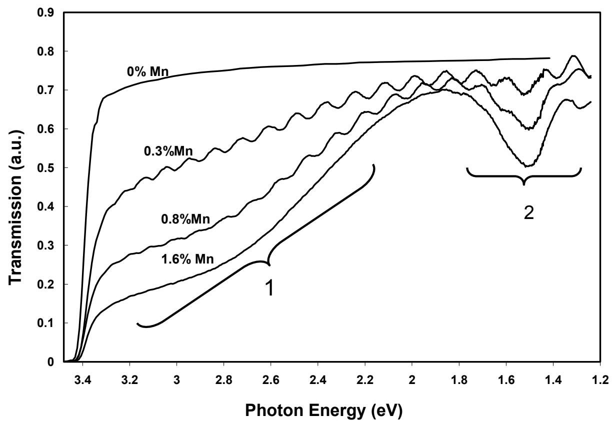

Fig. 1: Optical transmission spectra of GaMnN as a function of Mn content. Absorption mechanisms 1 and 2 scale with the Mn content of the films.

In the case of unintentionally doped GaN typical band edge absorption at 3.4eV is observed with no other features to note (interference fringes have been subtracted from this spectrum for clarity). In comparison to undoped GaN, two additional transitions are observed in the transmission spectra of the GaMnN films shown in Fig. 1. The first transition at 1.5 eV corresponds to the transition of carriers from the valence band of GaN to the Mn level; the second absoption band starting at 2.1eV is attributed to the transition of carriers from the Mn level to the conduction band of GaN. These two transitions will be referred to as transition 1 and transition 2, respectively. The transmission spectra for GaMnN films shows interference for low concentrations of Mn, however fringing disappears for higher concentrations due to surface roughening. Transition 1 is sharp and the absorption increases with increasing Mn concentration. Moreover it is observed that the width of this absorption band scales with the concentration of Mn, indicating that the Mn band widens as the concentration is increased. The full width at half maximum (FWHM) of the three absorption bands corresponding to increasing Mn contents are 107, 178 and 197meV, respectively.

Transition 2 has an onset of 2.1eV and extends up to the bandedge of GaN. The broad nature of this absorption band is attributed to transitions from the Mn *3d* band to various deep levels in the bandgap of GaMnN.

The effect of GaMnN film thickness on the transmission spectra was also investigated by growing 0.7µm, 0.94µm and 1.4µm thick films with the same concentration of 0.3 atom% Mn. Absorption at both transition 1 and 2 scales with the thickness of the films due to the higher absorption volume offered by thicker films. Interference fringes observed on the transmission spectrum also decreases with film thickness due to increased surface roughening.

Ferromagnetic properties in the GaMnN system were found to be a function of the Fermi level in the crystal. Recent results indicate that manipulation of the Fermi level by co-doping GaMnN with silicon or magnesium results in a reduction in the saturation magnetization (M. J. Reed et al. 2005). Figure 2 shows the saturation magnetization of the GaMnN films as they are doped with silicon and magnesium. These results are explained based on the occupancy of the Mn *3d* band formed as a result of doping GaN with Mn. Saturation magnetization is highest when the Mn *3d* band is partially occupied with carriers.

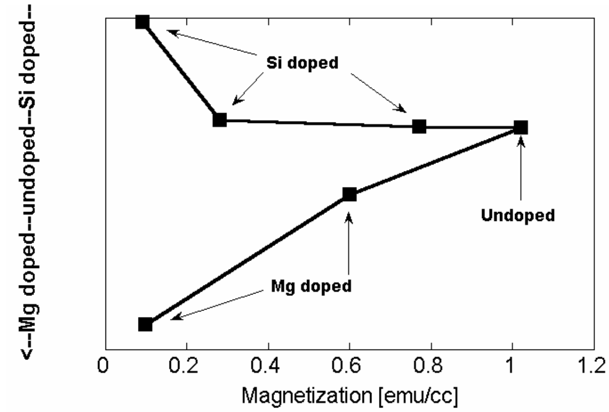

Fig.2: Saturation magnetization as a function of Si or Mg doping.

Depleting or filling this band by changing the Fermi level in the crystal via co-doping reduces the magnetization. Transmission measurements were performed on the undoped, Si doped, and Mg doped GaMnN films for which magnetization data is given in Fig. 2 to investigate the position of the Fermi level in the crystal as a function of doping.

Transmission measurements on samples which are unintentionally doped, lightly doped ($1x10^{18}/cm^3$) and heavily doped ($2x10^{20}/cm^3$) with silicon are performed. The saturation magnetizations of these samples are 0.9, 0.77 and 0.29 emu/cm^3 respectively. GaMnN films show ferromagnetic ordering when the Fermi level runs through the Mn band. When the Fermi level is raised above the Mn band, saturation magnetization decreases significantly. Figure 3 shows the transmission spectrum of these three samples and an unintentionally doped GaN sample as a reference.

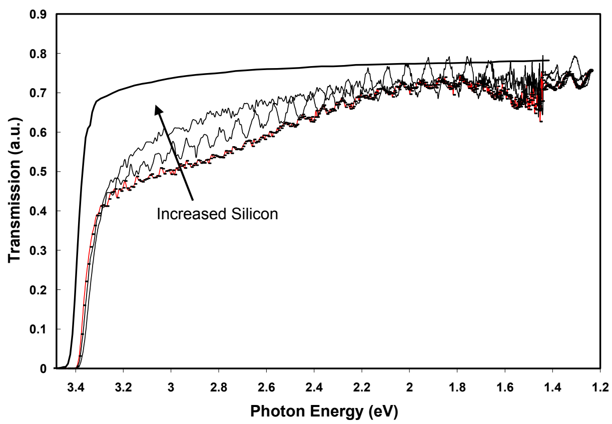

Fig. 3: Transmission spectra as a function of Si doping in GaMnN.

It is observed that as the silicon concentration is increased, the absorption band at 1.5 eV gradually becomes weaker and eventually disappears, indicating that the Mn band is occupied as more silicon is added. As a result, there are no empty states in the Mn energy band to which carriers in the valence band may be excited. This verifies that the decrease in the saturation magnetization accompanying co-doping of GaMnN with Si is a result the Fermi energy being well above the Mn acceptor level.

Figure 4 shows the effect of increasing Mg concentration in GaMnN films on the transmission spectra.

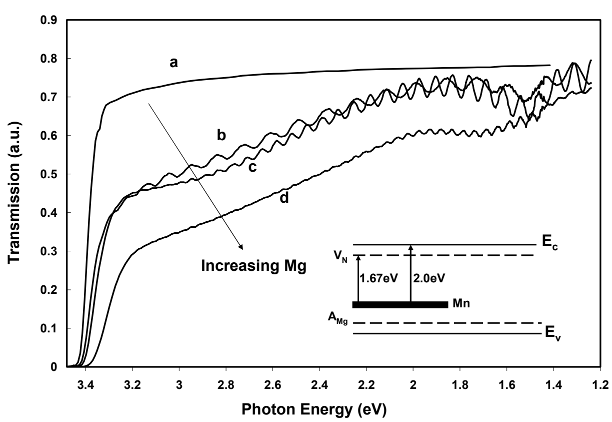

Fig.4: Optical transmission spectra as a function of Mg doping in GaMnN.

It can be observed that for low concentrations of Mg (curves b and c) the absorption band at 1.5 eV becomes stronger with increasing Mg concentration. The addition of Mg provides acceptor states to which carriers in the Mn band thermalize. As a result, the Mn band is depleted of carriers and excitation of electrons from the valence band into Mn acceptor states becomes more efficient. As the Mg concentration is increased further, the absorption band shifts to higher energy as seen in Fig. 4 curve d. This is attributed to the formation of nitrogen vacancies due to growth instabilities, which in turn provide donors that eventually thermalize into Mn levels. This shifted absorption band corresponds to the excitation of carriers from the Mn level to the empty donor levels created by nitrogen vacancies as shown in the inset of Fig. 4.

4. CONCLUSION

Transmission measurements of GaMnN samples co-doped with silicon and magnesium indicate that the Fermi level position is effectively manipulated by co-doping. Since the saturation magnetization of GaMnN films has been found to depend on the Fermi level, the use of transmission measurements is an excellent and rapid means of probing the Fermi level in GaMnN crystals and assessing the potential for ferromagnetic ordering in these samples. This allows more rapid optimization of deposition conditions used to produce these dilute magnetic semiconductors which have potential application in room temperature spintronic devices.

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