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USING DISPERSION THEORY TO INTERPRET THE POLARIZATION PROPERTIES OF THE BAND-EDGE EMISSION IN GaN CRYSTALS

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In this paper, we report the polarization properties of the near band-edge emission in GaN crystal wafers. When excitation intensity increases in the range of 0–0.1 MW cm^{-2}, the polarization of band-edge emissions increase quadratically, then, a saturation appears.

These phenomena can be explained by the dispersion theory of exciton–exciton interaction.

1. INTRODUCTION

IT IS WELL-KNOWN that, uniaxial crystal GaN is a direct gap semiconductor, a large amount of works on this promising material for luminescence and laser devices are aiming at the near band-edge emission [1–3].

In our previous papers [4, 5], we proposed the dispersion concept of exciton–exciton interaction. According to dispersion theory, in uniaxial crystal, the dispersion effect between bound excitons is unisotropic, the *C*-axis is the preferential direction. This property should make the luminescence polarization of bound excitons increase with the excitation intensity quadratically.

In this paper, the luminescence polarization properties of bound excitons (I_1 and I_3 lines) in uniaxial crystal GaN were studied carefully at the temperature of 77 K. In the excitation range of 0–0.1 MW/cm^{-2}, we found that, the polarization of band-edge emission I_1 and I_3 increased with I_{ex} quadratically at first, and then, a saturation appeared. These experiment facts support the dispersion theory of exciton–exciton interaction.

2. SAMPLES AND EXPERIMENTAL SET-UP

Samples used in the experiment were GaN single crystal wafers, which were grown by the vapour phase epitaxy method on sapphire substrates oriented in (0 0 0 1) direction [6]. There were two kinds of samples, one of them was unintentionally doped GaN crystal wafers, typical sample was labelled by GaN – 15 #, another was Zinc-doped GaN crystal wafers, typical was labelled by GaN – 16 #. The thickness of GaN crystal wafers was about 8 μm, the electron concentration of unintentionally doped GaN crystal wafers was about 10^{18} cm^{-3} at room temperature determined by Hall measurement.

Experimental work was finished on HEPLSM (high excitation photoluminescence spectrum measurement) system of Transient Solid Spectrum Laboratory, Changchun Institute of Physics, C.A.S.

Samples were held in the cryostat ESR – 900 and excited by a light beam, whose wavelength of 308 nm comes from an excimer laser EMG – 102, with a set of neutral attenuators, I_{ex} can be varied in the range of 0–100 MW cm^{-2} easily. A polarizer was put in before a grating double monochromator to pick out the luminescence intensity polarized in parallel or perpendicular to *C*-axis of GaN crystals respectively. After the monochromator, there was a photomultiplier C – 31034 with semiconductor cooling system to measure luminescence signal, and then, recorded by $X - Y$ recorder.

3. EXPERIMENT RESULTS

3.1. *Emission spectra*

The near band-edge emission spectra of GaN crystal wafers under proper excitation intensity I_{ex} were shown in Figs. 1 and 2.

There were three emission peaks that could be distinguished apparently in Fig. 1 and 2, which were situated at 3.466, 3.353, and 3.313 eV respectively. Compare GaN – 15 # and GaN – 16 #, one can see that, as a result of Zn-doping, the intensity of 3.466 eV line decreased and the other two lines (3.353 and 3.313 eV) increased. When I_{ex} increased, we found that, the peak wavelength of 3.313 eV line shifted to violent side. Combining these points with the temperature effects, we labelled these three peaks as I_1, I_3 and I_4

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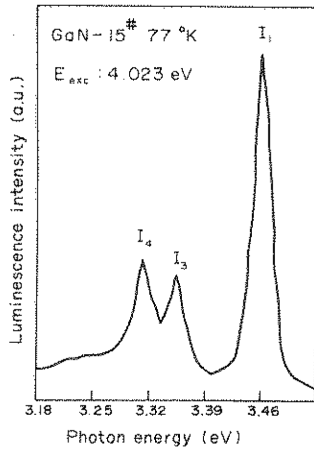

Fig. 1. The emission spectrum of GaN-15#.

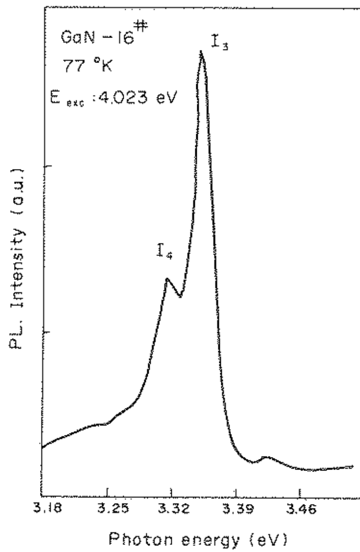

Fig. 2. The emission spectrum of GaN-16#.

line respectively, see [3]. In [3], I_1 line was due to neutral donor center bound excitons (BED°), I_4 was due to $D-A$ pairs, I_3 was due to neutral acceptor center bound excitons (BEA°). (Whether I_3 is due to the recombination of BEA°'s or isoelectron center bound excitons, need to be identified further, but this does not affect the basic idea of the paper).

3.2. *Experiment results of luminescence polarization*

At first, we define the luminescence polarization P as follows:

$$P = I_{\parallel}/I_{\perp}, \tag{1}$$

where, $I_{\parallel}$ and $I_{\perp}$ are the luminescence intensity parallel and perpendicular to C-axis of GaN crystal wafers respectively, see Fig. 3.

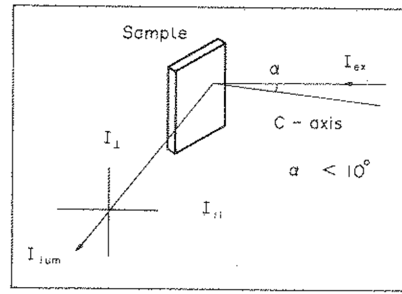

Fig. 3. The optical scattering plane of the experiment.

In experiments, we found that P has different values under different I_{ex} see Figs. 4 and 5.

From Figs. 4 and 5, some important points can be figured as follows:

(1) The near band-edge emission of GaN crystal wafers show polarization character, the preferential direction is C-axis.

(2) P values of I_1 and I_3 lines increase along with I_{ex} during the excitation range of 0–0.1 MW cm^{-2}.

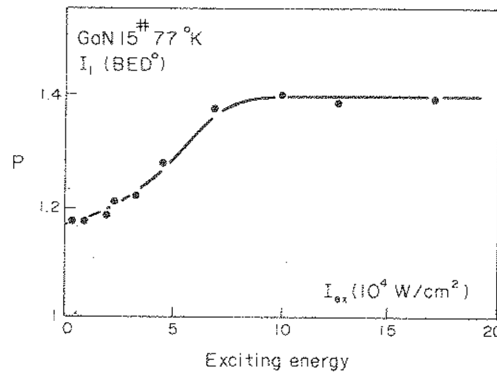

Fig. 4. P values of I_1 line under various I_{ex}'s.

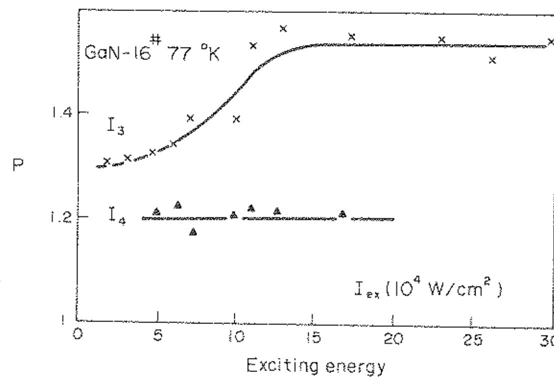

Fig. 5. P values of I_3 under various I_{ex}'s.

(3) When I_{ex} is about $0.1\,\text{MW cm}^{-2}$, the luminescence polarizations of I_1 and I_3 lines show saturation phenomena.

(4) P value of I_4 line do not change with I_{ex}.

4. ANALYSIS OF EXPERIMENT RESULT

4.1. *Dispersion theory*

The exciton has a polarization $\mathbf{p} \cdot \exp(iK \cdot R)$ associated with it. Like in neutral atom, the transient dipole–dipole interaction resulting in the Van der Waals Force which was known as Loudon dispersion force. When two excitons approach each other, the transient dipole–dipole interaction resulting the dispersion effect of exciton–exciton interaction also. We only cite some main ideas and formula from [4] and [5].

For example, we consider BED^0 case, $\mathbf{p}$ shows spherical-like distribution, hence BED^0 is a good sample to study the dispersion effect of exciton–exciton interaction. Suppose there are two BED^0's localized at $\mathbf{r}_1$ and $\mathbf{r}_2$, labelled by $\mathbf{p}_1$ and $\mathbf{p}_2$ respectively, $\mathbf{p}_1$ will induce a transient electrical field at $\mathbf{r}_2$, which will polarize $\mathbf{p}_2$ or make $\mathbf{p}_2$ larger, enlarged $\mathbf{p}_2$ will repolarize $\mathbf{p}_1$ with more power.

The interaction is supposed fast enough and then, both $\mathbf{p}_1$ and $\mathbf{p}_2$ get an increased quantity $\mathbf{M}$. The detailed calculation shows that:

$$M = (2)^{1/2}|\mathbf{p}|^3/3\epsilon R^3 \hbar w_0, \quad (2)$$

where, ϵ stands for the dielectrical function of GaN crystal, $\hbar w_0$ stands for the energy of BED^0, $\mathbf{R}$ stands for the nearest distance between BED^0 and other excitons (including free exciton, BED^0, BEA^0, or other kind of bound excitons):

$$\mathbf{R} = \mathbf{r}_2 - \mathbf{r}_1.$$

As we know, GaN is an uniaxial crystal, it has an unisotropic crystal field. We define a parameter "a" as an unisotropic parameter:

$$a = \epsilon_{\parallel}/\epsilon_{\perp} > 1, \quad (3)$$

where, ϵ stands for the dielectric function of GaN crystal, "$\parallel$" and "$\perp$" stand for the direction parallel and perpendicular to C-axis.

If one allows for the unisotropic structive effects of valence band in uniaxial crystals, the value of "a" would be larger than the value calculated by formulae (3).

The unisotropic crystal field should affect the electric distribution, make the transient dipole moment $\mathbf{p}$ not spherical but elliptical, that is, the parallel component of $\mathbf{p}$ would be larger than the perpendicular one:

$$p_{\parallel} = aP_{\perp} > p_{\perp}. \quad (4)$$

According to formula (2), (3) and (4), one can deduce a relation:

$$M_{\parallel} = a^4 M_{\perp} > M_{\perp}, \quad (5)$$

where, $M_{\parallel}$ and $M_{\perp}$ stand for the induced transient dipole moment parallel and perpendicular to C-axis, respectively.

Formulae (5) show that, in uniaxial crystal, such as GaN, the dispersion effect has the unisotropic character.

Allowing for Fermi relation between dipole moment and luminescence intensity, from formulae (1), can get the following relation:

$$P = I_{\parallel}/I_{\perp} = (P_{\parallel} + M_{\parallel})^2/(P_{\perp} + M_{\perp})^2, \quad (6)$$

where, $I_{\parallel}$ and $I_{\perp}$ are the luminescence intensities parallel and perpendicular to C-axis.

Approximately, consider the optical absorbtion process as a single molecular approach, write down the relation between I_{ex} and the density N of BED^0, and the relation between N and the nearest distance R:

$$I_{ex} \cdot f/\hbar w \cdot \text{De}\, f = N/\tau, \quad (7)$$

$$R^{-3} = 4\pi N/3, \quad (8)$$

where f: the oscillator strength of BED, τ: the lifetime of BED^0, De f: the effective absorbtion depth of GaN crystal wafers, $\hbar w$: exciting photon energy.

From formula (3), (6), (7) and (8), one can get a relation:

$$P = (A + B \cdot I_{ex})^2, \quad (9)$$

where, A: a constant relevant to the initial polarization of BED^0, B: a constant relevant to the dispersion effect of BED^0 by another BED^0 or other excitons.

Detailed deducement shows that:

$$A = a, \quad (10)$$

$$B = 4\pi \cdot a \cdot f \cdot \tau \cdot |\mathbf{p}|^2/9\epsilon\hbar w_0 \cdot \hbar w \cdot \text{De}\, f. \quad (11)$$

Take BED^0 as a sample in GaN crystal, unisotropic parameter $a \doteq 1$, oscillator strength $f \doteq 1$, lifetime $\tau \doteq 10^{-9}$ second, transient dipole moment $|\mathbf{p}| \doteq 17\,\text{eÅ}$ (the radius of exciton in GaN is about 17 Å), dielectric costands $\epsilon \doteq 6\epsilon_0$, the energy of BED^0 $\hbar w_0 \doteq 3.400\,\text{eV}$, exciting photon energy $\hbar w \doteq 4.023\,\text{eV}$, the effective absorbtion depth De $f \doteq 1\,\mu\text{m}$, one can calculate the values of A and B:

$$A \doteq 1 \quad (12)$$

$$B = 10^{-6}\,(\text{cm}^2\,\text{W}^{-1}) \quad (13)$$

4.2 *Experiment result analysis*

Figures 5 and 6 show the relation between $(P)^{1/2}$ and I_{ex}'s.

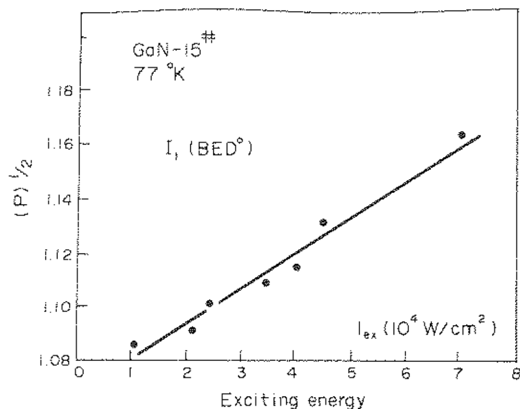

Fig. 6. $(P)^{1/2} - I_{ex}$ relation of I_1 line.

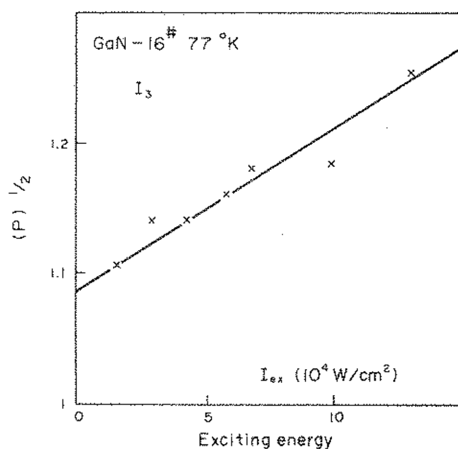

Fig. 7. $(P)^{1/2} - I_{ex}$ relation of I_3 line.

From Fig. 6 one can know that, for I_1 line, $A \doteq 1.07$, $B \doteq 1.30 \cdot 10^{-6}$ ($cm^2\,W^{-1}$). From Fig. 7 one can know that for line I, $A \doteq 1.09$, $B \doteq 1.20 \cdot 10^{-6}$ ($cm^2\,W^{-1}$). All of the experimental values can suit well the theoretical calculating values.

When I_{ex} is over about $0.1\,MW\,cm^{-2}$, the free exciton density will be about $10^{18}\,cm^{-3}$, various kinds of bound centers are occupied by excitons or in another words, the bound excitons are saturated. After that, when I_{ex} increases further, the distance of the nearest neighbors does not apparently shorten. This is the reason of the saturation phenomena in luminescence polarization of I_1 and I_3 lines appeared in Figs. 4 and 5.

Since I_4 line is not relevent to excitons, the dispersion concept does not suit it, and it just shows the initial polarization property caused by the unisotropic crystal field (both the dielectric function and the band structive effects) of GaN crystal wafers.

5. CONCLUSIONS

From the above analyses, one can make some conclusions as follows:

The near band-edge emissions of uniaxial crystal GaN including I_1, I_3 and I_4 show initial polarization properties, *C*-axis is the preferential direction.

The luminescence polarizations of emission lines relevent to bound excitons increase quadratically with I_{ex} in the excitation range of 0–0.1 $MW\,cm^{-2}$. Meanwhile, polarization of I_4 line does not change with I_{ex}. These experimental results gives support to the dispersion theory of exciton–exciton interaction strongly.

Because the density of bound excitons is limited by the bound centers, when I_{ex} is about $0.1\,MW\,cm^{-2}$, the saturation phenomena of luminescence polarization of bound excitons appeared.

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