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Influence of point defects on optical properties of GaN-based materials by first principle study

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ABSTRACT

Although GaN-based materials have been successfully utilized in optoelectronic and electronic devices, some important physical issues have not been fully understood, for example, why they can have high luminous efficiency even with high density of point defects, and what are the origins of yellow luminescence (YL) in their photoluminescence (PL) spectra. In this paper, the influence of point defects on GaN-based materials are investigated by first principle calculation. It is revealed that vacancy defects in GaN-based materials only make the effective masses of electrons and holes increase, consequently they have no significant contribution on the non-radiative recombination. What is more, C—O complex interstitial impurities or Ga interstitial impurities are most likely to lead to YL in PL spectra of GaN-based materials.

1. Introduction

Due to their superior intrinsic properties, GaN-based materials have attracted considerable attentions, and have been successfully utilized in optoelectronic and electronic devices such as blue light emitting diodes (LEDs) and high electron mobility transistors (HEMTs) [1]. Currently GaN-based materials grown on sapphire by metal-organic vapor phase epitaxy (MOVPE) still have a very high density of point defects ($V_{Ga} > 10^{19}\,cm^{-3}$) [2]. In fact, the radiative recombination efficiency of the InGaN/GaN quantum wells can be up to 80–90% [3] even with such high density of point defects. Previous studies showed that the carrier localization [4] was a possible reason of this phenomenon, however this theory cannot explain the phenomenon in bulk GaN materials.

On the other hand, GaN-based materials are well known for having several deep-level states, which were supposed to be related to the yellow luminescence(YL) in photoluminescence (PL) spectra [5,6]. Although it has been investigated extensively over the last two decades, the cause of YL is still elusive. The threading and screw dislocations were considered to have no significant correlation with YL [7,8]. In an early experiment by Ogino and Aoki [9], they believed YL in GaN was caused by a complex involving Ga vacancies (V_{Ga}) and C antisites (C_N), while in contrast some other studies suggested that V_{Ga} and C_N were not the reasons of the formation of GaN YL [10–12]. The strong correlation between the YL intensity and carbon concentration has been widely observed [13–17]. Demchenko et al. recently performed a hybrid functional study on the excited state with different point defects [17], they found that the C_N—O_N complex can explain the formation of YL better [18]. However, this explanation is still contentious [19]. Recently, the relative position of the deep level defects caused by O impurity atoms was determined by the double laser excited PL in the GaN bandgap [21]. In short, the cause of the YL in GaN-based materials is still a large controversy.

In this paper, we focus on the effect of the point defects including vacancies and impurities on the optical properties of GaN-based materials with first principles calculation. It is found that the vacancy defects will not introduce deep level in the forbidden band. Their influence is mainly changing the curvature of conduction or valence band, namely effective masses of electrons or holes increased. Consequently, the vacancy defects would not contribute to the non-radiative recombination and the formation of the YL. On the other hand, our study on the impurity point defects shows that C—O interstitial impurities or Ga interstitial impurities are likely to be the main cause of the YL in GaN-based materials.

2. Calculation details

The calculations in this paper were based on density functional theory(DFT) and done by Vienna Ab initio Simulation Package

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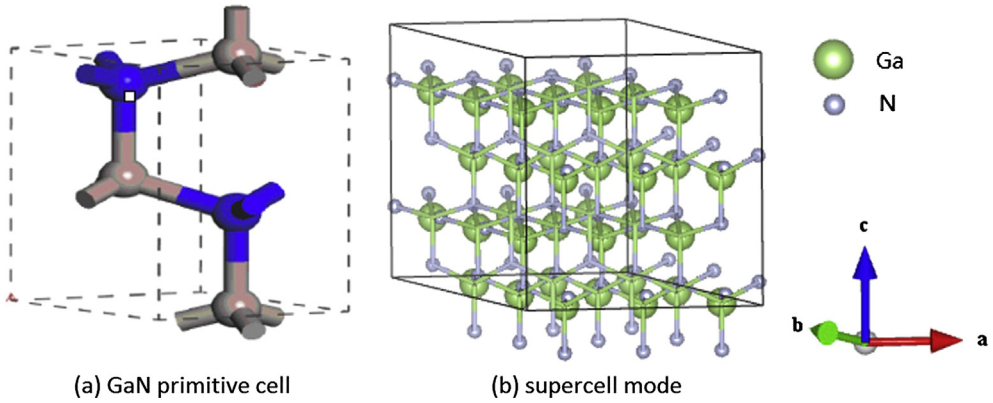

Fig. 1. Calculation model of GaN, (a) primitive cell and (b) ideal 72-atom supercell.

Table 1
The formation energies of point defects in GaN.

Point defects	Formation energy (eV)
V_N	3.03
V_{Ga}	4.36
N_{int}	5.75
Ga_{int}	6.28
N_{Ga}	7.04
Ga_N	7.84

(VASP) [22–27] with the GGA functional of Perdew, Burke, and Ernzerh of hybrid functional [28] as implemented in. The Ga 3d electrons were treated as core states within the projector-augmented wave method. The defects calculation was performed with a 400 eV plane-wave cutoff. Before starting the calculations, bulk GaN crystal structure was optimized with full relaxation. The convergence was set to energy change below 2×10^{-6} eV/atom, force less than 0.005 eV/nm, the convergence tolerance of a single

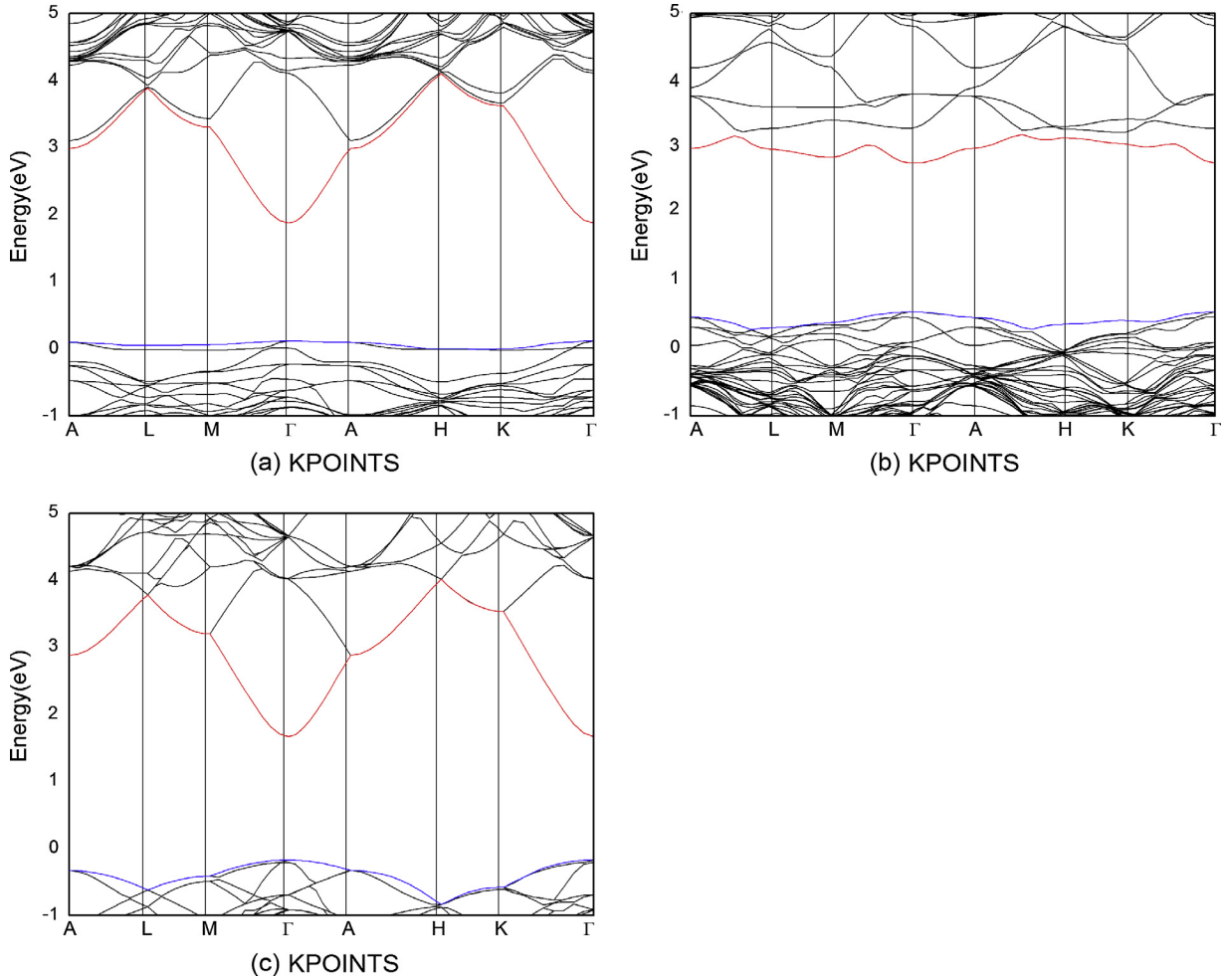

Fig. 2. The calculated energy band structures of GaN materials with (a) Ga vacancy and with (b) N vacancy and (c) ideal GaN crystal using the 72-atom supercell model.