Magnetic-Mechanical-Electrical-Optical Coupling Effects in GaN-Based LED/Rare-Earth Terfenol-D Structures

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Multifunctional micro/nano devices and systems are used in important applications in smart electronics for health care, human-machine interfacing, infrastructure monitoring and security. Among all of the known materials properties, magneto-optics and electro-optics, magneto-electrics and magneto-mechanics, piezotronics and piezo-phototronics, are the typical results of coupling between physical properties, with potential for sensing, manipulating, communicating and actuating/responding.[1,2] However, experimentally realizing these couplings is challenging because one material may not simultaneously exhibit superior multi-properties in responding to electrical, optical, mechanical and magnetic excitations. In this research, modulation and realization of multi-field coupling effects is realized in GaN based LED/Tefenol-D multilayer structure, which can broaden the recognition of new phenomena, unprecedented multifunctionality, and multiplex system integration.

Piezoelectricity is a well-known effect that a mechanical stimulation is converted into an electrical response or vice versa. Recently, the piezotronics effect of wurtzite semiconductors (e.g., GaN, InN, ZnO and ZnS) has attracted intensive attentions by conjunction of the piezoelectric effect and the electronic transport process by using the polarization charge induced interface potential as a “gate” voltage for controlling carrier transport.[3–5] Further more, the piezo-phototronic effect has been developed by using piezoelectric polarization charges to tune the optoelectronic processes in solar cells and n-ZnO nanowire/p-GaN light-emitting diodes.[6–10] Besides, in multiferroic magnetoelectric materials, the magnetoelectric coupling effect is a result of the magnetostrictive effect (magnetic-mechanical coupling) in the magnetic phase and the piezoelectric effect (mechanical-electrical coupling) in the piezoelectric phase.[11,12] It is well known that the perovskite structured PZT materials are the best performance piezoelectric materials, however, they are seldom used for either optoelectronic or magnetic applications because of the limited performances. Inversely, the excellent optoelectronic semiconductor materials seldom possess magnetic property. Therefore, there is not a single one-material that can be effective for exploring the multi-coupling effects among magnetic, mechanical, electric and optical interactions.

In this work, a multi-field coupling structure is designed and investigated, which combines GaN-based optoelectronic devices and rare-earth Terfenol-D ($Tb_xDy_{(1-x)}Fe$ alloys). As excellent optoelectronic semiconductors, nonmagnetic III-nitride materials have become the most important building blocks for LEDs and laser diodes operating in visible to ultraviolet range of the optical spectrum.[13,14] Owing to the piezoelectric polarization effect caused by the lattice mismatch among wurtzite GaN-based multilayers, a high internal piezoelectric field along the c-axis of InGaN/GaN QWs has a significant impact on the optoelectronic performances of GaN-based LED devices. As a rare-earth alloy, the giant magnetostriction effect of Terfenol-D can offer large tensile strain and force strength to relax the compressive stress in InGaN/GaN multiple quantum well (MQW) LEDs on sapphire substrates. The well-designed structure can bring together these two disparate material types to create coupling among multifunctional properties. The abundant coupling effects and multifunctionalities among magnetics, mechanics, electrics and optics will be fully investigated by a combination of nonmagnetic GaN-based piezoelectronic optoelectronic characteristics and giant magnetomechanical property of Terfenol-D. A few potential new areas of studies are proposed.

Figure 1 shows the GaN based LED/rare earth Terfenol-D multilayer structure. The blue InGaN/GaN MQW LED was grown on c-plane sapphire substrates. From bottom to top, the LED consisted of 2 µm undoped GaN buffer layer, 3 µm n-type GaN layer, five periods of $In_{0.18}Ga_{0.82}N$/GaN (3 nm/12 nm) quantum well active layers, 60 nm p-type $Al_{0.15}Ga_{0.85}N$ electron blocking layer and 100 nm p-type Mg-doped GaN capper layer. The bulk Terfenol-D and InGaN/GaN MQW LED chip were tightly bonded using hard epoxy resins. The magnetization easy axis of Terfenol-D is along its longitudinal direction. In optoelectronic measurements, current was applied along the thickness direction of the LED and light was collected from the top of the multilayer structure. All of our measurements were made at room temperature.

In **Figure 2**, multi-property coupling effects among magnetics, mechanics, electrics and optics were summarized, including two-way coupling effects (magnetoelectrics, magnetostriction, magnetophotonics, optoelectronics, piezotronics and piezo-photonics), and three-way coupling effects (magneto-optoelectronics, piezo-magnetotronics and piezo-phototronics).[15] In this multifield coupling system, magnetoelectrics is an effect of inducing magnetic/electric polarization by applying

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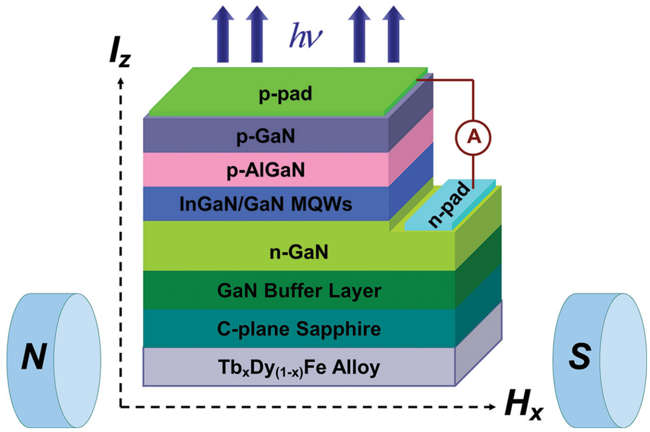

Figure 1. The EL spectrum measurement setup of the MMEO multilayer structure under magnetic field. The as-mentioned multilayer structure consists of $Tb_xDy_{(1-x)}Fe$, c-plane sapphire, GaN buffer layer, n-GaN, $In_{0.18}Ga_{0.82}N$/GaN MQWs, p-AlGaN and p-GaN from bottom to top.

an external electric/magnetic field, which was conjectured by P. Curie in 1894 and then observed by P. Debye in 1926;[16] Magnetoelastics is a property of ferromagnetic materials that are able to change their shape or dimensions imposed by magnetization. It was first identified in iron by J. Joule in 1842;[17] Magnetophotonics is based upon Faraday effect that polarization of optical wave can be rotated during the wave transmitting through a magneto-optical medium for the optical manipulation;[18] Optoelectronics is electrical-to-optical or optical-to-electrical interactions in semiconductor electronic devices, such as photon detector, solar cell and LED; Piezotronics, first reported in 2007, uses the stress/strain driven piezopotential as a "gate" voltage to tune/control charge carrier transport at a contact or junction;[19] Piezo-photonics is an effect of optical-mechanical conversion in micro-opto-electro-mechanical systems, which has application insensing or manipulating optical signals by using piezoelectric polarization field. In addition, magneto-optoelectronics, piezo-magnetotronics and piezo-phototronics are much more complicated, which involve three-way couplings among magnetics-electrics-optics, magnetics-electrics-mechanics and mechanics-electrics-optics, respectively. Hereinto, piezo-phototronics is to use the piezocharges to control the carrier generation, transport, separation and/or recombination to improve the performance of optoelectronic devices.[20]

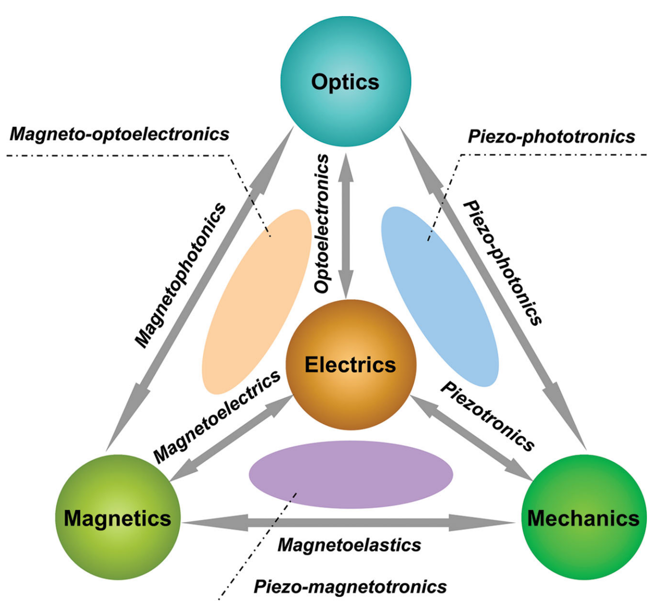

Figure 2. The schematic diagram of multi-field coupling effects among physical quantities of magnetics, mechanics, electrics and optics, including two-way coupling effects (magnetophotonics, magnetoelectrics, magnetostriction, optoelectronics, piezotronics and piezophotonics), and three-way coupling effects (magneto-optoelectronics, piezo-magnetotronics and piezo-phototronics).

In our multilayer structure, GaN has the unique piezoelectric and semiconducting properties; Terfenol-D has magnetoelastic or so-called magnetostrictive effect; InGaN/GaN MQW has tunable optoelectronic effect. Furthermore, the combination of two or more effects mentioned above can generate multi-field couplings as shown in Figure 2. For example, piezotronic effect, originating from the combination of piezoelectric and tunable semiconductor properties, is present in InGaN/GaN MQW structure (Figure S2, supporting information); magnetoelectric effect, the coupling of piezoelectric and magnetostrictive effect, occurs in the InGaN/GaN MQW/Terfenol-D multilayer structure; piezo-magnetotronic coupling effect (the combination of piezotronics and magnetoelastic effect) exhibits in the whole multilayer structure. In **Figure 3**(a), the current-voltage (I–V) characteristics of our multilayer structure were measured under various magnetic fields. The insert graph of Figure 3(a) shows that, when applied a constant voltage to multilayer structure, the induced current reduced with the increase of magnetic field. Along the [0001]-axis orientation of InGaN/GaN MQW epilayers, negative and positive piezoelectric charges presented at p-GaN epilayer and n-GaN epilayer sides, respectively, under tensile magnetostrictive strain induced by Terfenol-D underneath. Because of the high-density free carrier screening effect, the piezoelectric polarization potentials at both sides of the LED device were reduced greatly. As a result, the equilibrium piezoelectric potentials induced a small reverse bias voltage, and the turn-on threshold voltage of InGaN/GaN LED device increased from 2.5 V at H = 0 mT to 2.52 V at H = 320 mT. The increase of turn-on threshold voltage further confirms the existence of magnetics-mechanics-electrics coupling effect.

In magnetics-mechanics-electrics coupling effect, both lattice mismatch strain (ε_1) and magnetostrictive strain (ε_H) determine piezoelectric polarization of InGaN well layer and GaN barrier layer. The [0001]-axis piezoelectric polarization (P_{pz}) induced by ε_1 and ε_H strains can be given by Equation (1):

$$\begin{cases} P_{pz} = e_{33}\varepsilon_3 + 2e_{31}(\varepsilon_1 - \varepsilon_H) \\ \varepsilon_3 = -2\varepsilon_1 C_{13}/C_{33} \\ P_{pz} = 2\varepsilon_1(e_{31} - e_{33}C_{13}/C_{33}) - 2e_{31}\varepsilon_H \end{cases} \quad (1)$$

where e_{33}, e_{31} and C_{33}, C_{13} denote the piezoelectric and elastic constants of nitride films, respectively.[21] Without the magnetic field modulation, $In_{0.18}Ga_{0.82}N$ well layer has a coherently compressive strain at around 1.95% and a piezoelectric polarization of 0.0281 C/m^2. For $In_{0.18}Ga_{0.82}N$/GaN MQW heterostructure, the difference in spontaneous polarization between adjacent

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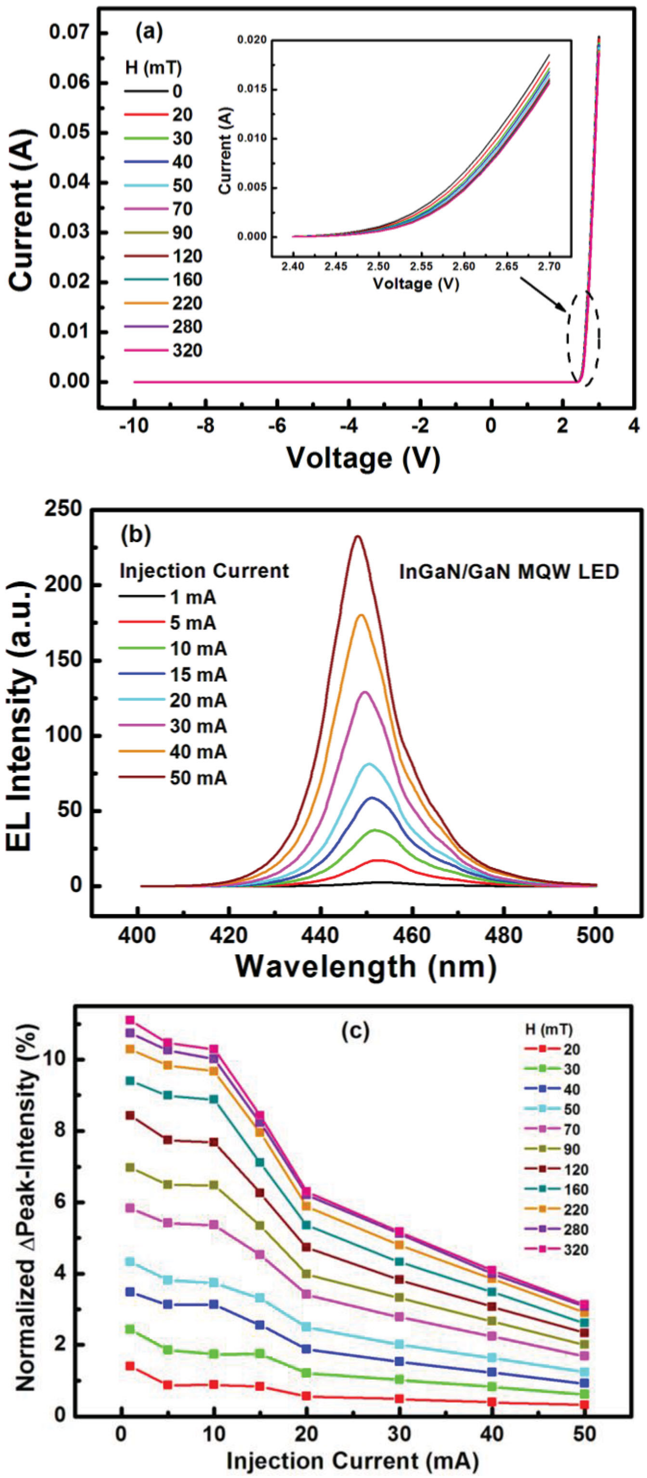

Figure 3. (a) The current-voltage (*I–V*) characteristic of the MMEO multilayer structure under the magnetic field from 0 to 320 mT. The insert graph enlarges up the device threshold region of rectifying *I–V* curves. (b) The EL spectra of our $In_{0.18}Ga_{0.82}N$/GaN MQW LED device under various injection currents. (c) The normalized EL peak-intensity change of the MMEO composite structure under various magnetic fields.

layers is much smaller, only -0.00054 C/m^2. Here, the piezoelectric polarization is a dominant factor, and its direction is opposite to that of the spontaneous polarization charge. When magnetostrictive strain from Terfenol-D increases with the increasing magnetic field, the total piezoelectric polarization in $In_{0.18}Ga_{0.82}N$ well layer will be partially reduced by magnetostrictive modulation. Such modulation of piezoelectric polarization at the $In_{0.18}Ga_{0.82}N$/GaN multilayer interfaces will influence the carrier transport, energy level transition, and recombination processes.

In addition to electronic properties, the nonmagnetic InGaN/GaN MQW LED also has tunable optoelectronic effect. When the electrons in n-GaN and the holes in p-GaN are injected into InGaN/GaN MQW region under a forward bias, the electrons in the conduction band will jump into the vacancy positions in the valence band simultaneously with the generation of photons. The EL spectra of the InGaN/GaN MQW LED were measured under various electrical injection conditions in Figure 3(b). As the injection current increased from 1 mA to 50 mA, the EL spectra showed remarkable luminescence enhancement because of the photo-electric conversion. In addition, a blue-shift of luminescence peak was observed with the increase of the injection current. It demonstrates that a strong internal piezoelectric field along polar [0001]-axis is caused by the lattice mismatch between InGaN and GaN thin-film layers. The internal piezoelectric field induces a spatial separation of electron and hole wave functions in InGaN QW regions, thereby reduces the recombination probability, affects carrier lifetime and decreases internal quantum efficiency.[22–24] It is widely considered as a limiting factor of optoelectronic performances in polar InGaN/GaN MQW LED devices. Furthermore, the optoelectronic effect in the InGaN/GaN MQW LED is able to combine with the inherent piezoelectric effect to generate piezo-phototronic effect. More detailed information about piezo-phototronic effect of the InGaN/GaN MQW LED can refer to Figure S3 of Supporting Information.

Besides the tunable optoelectronic effects, the optical properties of our magnetic-mechanical-electrical-optical (MMEO) multilayer structure can also be modulated by the magnetic-force controlled piezoelectric charges. This coupling is an indirect magneto-optoelectronics effect. As we know, the external quantum efficiency of LED device is expressed as the photons emitted per electrons injected in. The changes of external quantum efficiency can be directly reflected by the EL intensity change under the same injection current (I). In order to investigate the effect of the magnetic field on the EL efficiency of InGaN/GaN MQW LED device, we normalized and defined the change of EL peak intensity (ΔPeak-Intensity) as $\frac{\Phi B - \Phi 0}{\Phi 0}$, where Φ_B and Φ_0 are the EL peak intensity with and without magnetic field, respectively. Under different injection currents passing through the InGaN/GaN MQW LED device, the changes of EL peak intensity as a function of magnetic fields are shown in Figure 3(c). A decline relationship is observed between the change of EL peak intensity and the injection current. It demonstrates that the magnetic-force induced polarization charges are screened by a lot of injected carriers under high current conditions. Thus, low injection current facilitates the EL enhancement.

In low magnetic field range (0–400 mT), the nonmagnetic GaN-based materials and LED devices have weak ability to tune their electronic and optical properties by magnetic field (Figure S1, Supporting Information). However, Terfenol-D has

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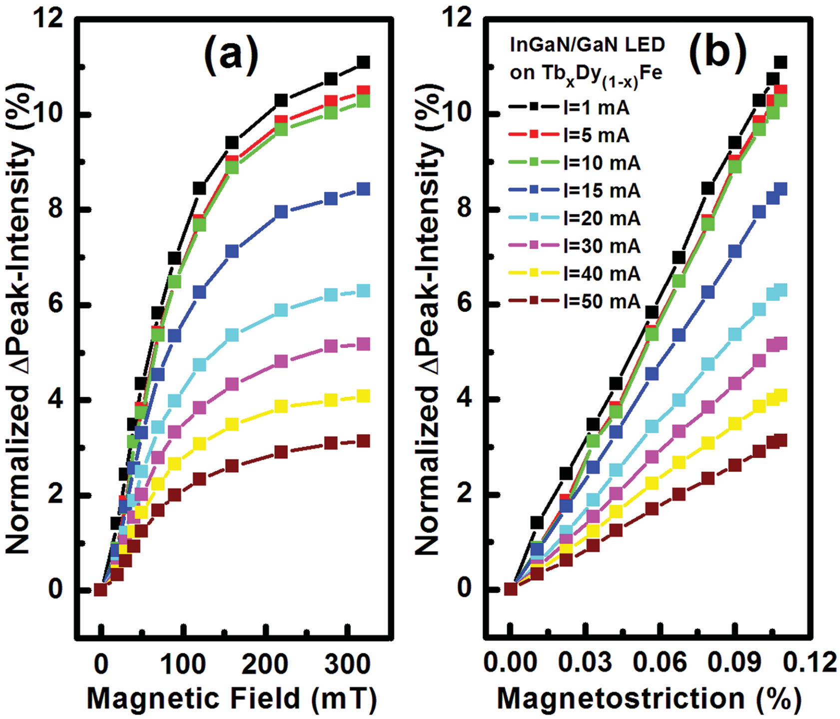

Figure 4. (a) and (b) are the normalized EL peak-intensity change of the MMEO multilayer composite as a function of magnetic field and magnetostriction, respectively.

excellent magnetoelastic property. It can be used to modulate electrical and optical modulation of InGaN/GaN MQW LED device. Figure S4 shows the magnetostriction (ε_H) of Terfenol-D as a function of external magnetic field at room temperature. As the magnetic field increased, the magnetostriction of Terfenol-D increased fast in low magnetic field range and saturated at high magnetic field. The maximum magnetostriction of 0.104% is reached at 320 mT. Therefore, the Terfenol-D can apply large tensile strain and/or stress to modulate piezoelectric polarization and optical properties of InGaN/GaN MQW LED by applied magnetic field.

The EL peak intensity change of our MMEO multilayer structure has also been investigated in terms of magnetic field and magnetostriction. As shown in **Figure** 4(a), the EL peak intensity obviously enhanced as the magnetic field increased. The value of ΔPeak-Intensity increased faster at low magnetic field and finally reached saturation at high magnetic field. This trend is the same as the change of magnetostriction of the Terfenol-D. On the other hand, a linear relationship between the change of EL peak intensity and the magnetostriction value of Terfenol-D is shown in Figure 4(b). At the maximum magnetostriction value of 0.104%, the ΔPeak-Intensity value reaches 11.09% and 3.13% at injection current of 1 mA and 50 mA, respectively. Therefore, the magnetics-electrics-optics coupling or magneto-optoelectronic effect of our multilayer structure is a combination of piezoelectric, magnetoelastic and phototronic effects.

To further investigate how the magnetic field tunes the piezoelectric polarization and carrier recombination process, band diagrams of the MMEO multilayer structure are calculated based on the carrier transport model using SiLENSe 5.4 software. Both spontaneous and piezoelectric polarization effects are taken into account. There exists a strong polarization field in [0001]-polarity MQW active region, which leads to the energy band bending. Because the Terfenol-D has tensile strain under magnetic field, it will cause partial relaxation of the intrinsic compressive strain accumulated in [0001]-polarity LED epilayers. The piezoelectric polarization charges at five-period $In_{0.18}Ga_{0.82}N$/GaN QW areas can be tuned by the magnitude of the magnetostrictive effect. The fundamental principle of the MMEO coupling effect is that magnetic-force induced polarization charges significantly modify the band structure at the InGaN/GaN multilayer interface as illustrated in **Figure 5**. The oblique triangular quantum wells without magnetic field in Figure 5(a) become more flat as magnetic-force induced polarization charges accumulate at multilayer interface in Figure 5(b). For clarity, we take the single QW (the fourth one in Figure 5(c) and (d)) as an example. The magnetic-force induced polarization charges can promote spatial overlapping of electron and hole wave functions, and thus increase their recombination probability. The energy level transition and radiative recombination processes between electrons and holes have been greatly improved. Therefore, it indicates that the optoelectronic property of the InGaN/GaN MQW LED device can be efficiently modulated by manipulating magnetostrictive and piezoelectric coupling effects, resulting in a high LED output.

In this study, an integrated structure for studying multi-field coupling is demonstrated by integrating nonmagnetic GaN-based optoelectronic semiconductors and Terfenol-D, using which, the MMEO coupling effects have been experimentally studied. It is shown that piezoelectric polarization and optical properties of InGaN/GaN LEDs can be modulated by the Terfenol-D. Under a low magnetic field (<400 mT), the giant magnetomechanical property of the Terfenol-D can offer magnetostriction to partially create a compressive strain along the [0001]-polarity direction of the InGaN/GaN MQW epilayers. The modulation of magnetic-force driven piezocharges in InGaN/GaN MQW region will influence the electrical and optical performances of the GaN-based LED device, which is a result of the MMEO coupling. By increasing the applied magnetic field, the electroluminescence (EL) peak intensity of InGaN/GaN MQW LED device was gradually enhanced. There exists a nearly linear relationship between its change in EL peak intensity and magnetostriction of Terfenol-D. This integrated multilayer structure proves the possibility of coupling among multi-fields. Our study shows the possibility of coupling among multi-fields effects, which could be of interest for fabricating functional devices in the fields of energy conversion, magnetic/optical imaging, high-density optical communication and information storage, smart sensing, and so on.

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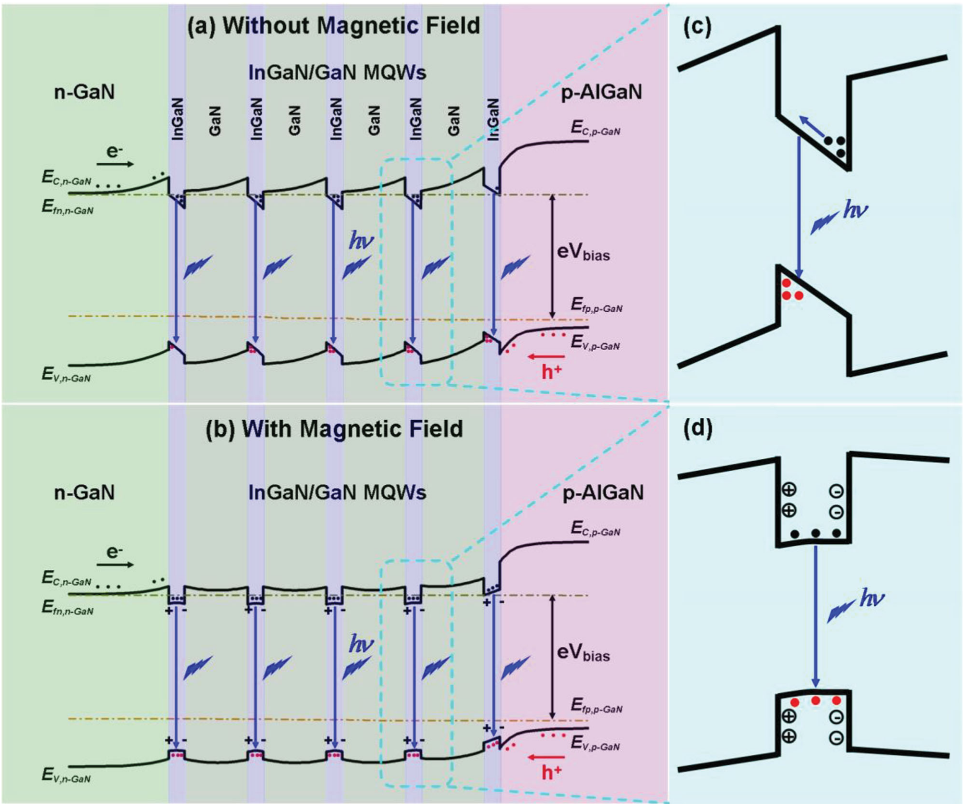

Figure 5. Energy band diagram of five-period InGaN/GaN QWs structure (a) with and (b) without magnetic field modulation. (c) and (d) gives the corresponding enlarged schematic diagram of the fourth QW in (a) and (b), respectively.

Experimental Section

The blue InGaN/GaN MQW LEDs were grown on c-plane sapphire substrates by low-pressure metal organic chemical vapor deposition (MOCVD) system, as shown in Figure 1. The gallium, indium and nitrogen sources were trimethylgallium (TMGa), trimethylindium (TMIn), and ammonia (NH_3), respectively. Biscyclopentadienyl magnesium (CP_2Mg) and silane (SiH_4) were used as the p-type and n-type doping sources, respectively. Planar GaN-based LED chips (250 μm × 250 μm) were fabricated as follows. First, active regions were defined by inductively coupled plasma dry etching down to n-type GaN layer. Then, Ti/Al/Ni/Au n-type and ITO p-type electrodes were deposited by using electron beam evaporation, respectively. After that, the bulk Terfenol-D and InGaN/GaN MQW LED chip were tightly bonded together using hard epoxy resins. Figure S5 showed our results have good reproducibility. Figure 1 presented the diagram of the setup for EL spectrum measurement of the device under magnetic field. Direct current supply and electrical measurement were carried out by using a Keithley 2400 source meter. The magnetic field was applied along longitudinal direction of Terfenol-D and measured by digital Gauss/Tesla meter. A coupled fiber with a grating spectrometer (Omni-λ 3008, Zolix Instruments) was used to collect and analyze the EL spectra under different magnetic fields.

Meanwhile, the energy band structure of InGaN/GaN MQWs simulation was calculated using the SiLENse 5.4 (STR inc.) modeling package. The Poisson equation was used for the electric potential, and one dimensional drift-diffusion transport equations were applied to study the electron and hole concentrations.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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