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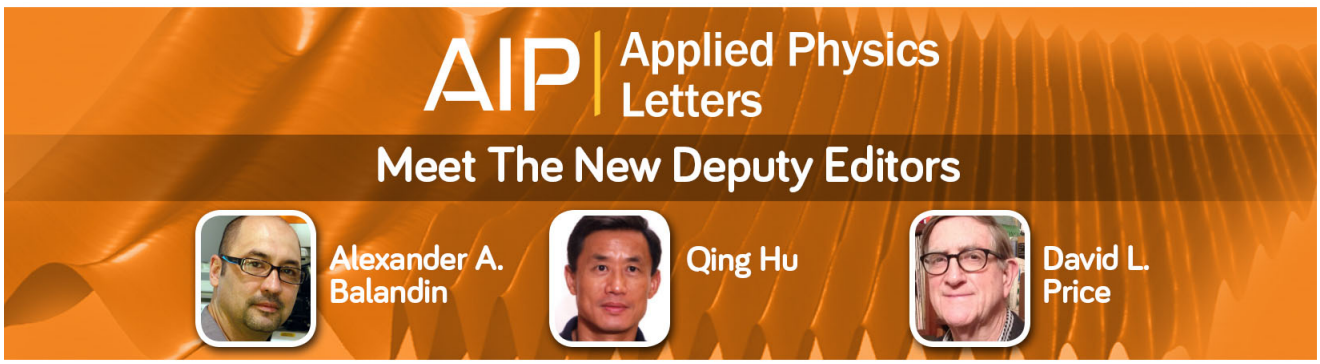

Looking for Auger signatures in III-nitride light emitters: A full-band Monte Carlo perspective

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Recent experiments of electron emission spectroscopy (EES) on III-nitride light-emitting diodes (LEDs) have shown a correlation between droop onset and hot electron emission at the cesiated surface of the LED *p*-cap. The observed hot electrons have been interpreted as a direct signature of Auger recombination in the LED active region, as highly energetic Auger-excited electrons would be collected in long-lived satellite valleys of the conduction band so that they would not decay on their journey to the surface across the highly doped *p*-contact layer. We discuss this interpretation by using a full-band Monte Carlo model based on first-principles electronic structure and lattice dynamics calculations. The results of our analysis suggest that Auger-excited electrons cannot be unambiguously detected in the LED structures used in the EES experiments. Additional experimental and simulative work are necessary to unravel the complex physics of GaN cesiated surfaces. [http://dx.doi.org/10.1063/1.4908154]

When, in 1922, Lise Meitner described radiationless transitions to explain the emission of electrons with signature energies as a corollary of an article on nuclear physics,[1] a remarkable discovery went unnoticed. The effect, known after Pierre Auger, who independently discovered it in 1923,[2] is now at the heart of the debate concerning droop, the decline of the internal quantum efficiency in GaN-based light-emitting diodes (LEDs) at high injection currents.[3–16]

Two notable recent experiments[12,13] have reported the direct observation of hot carriers excited by Auger recombination in the active region of InGaN/GaN light emitters. Binder *et al.*[13] optically pumped into droop regime a test structure composed of alternating ultraviolet (UV) and green quantum wells (QWs) using a blue high-power laser source. The observed UV luminescence emission was ascribed to hot electrons escaping the green QWs by Auger transitions and subsequently captured by the UV QWs, loss channels caused by current injection and leakage being ruled out by the optical injection. Iveland *et al.*[12] studied the energy distribution curves (EDCs) of the electrons emitted from the cesiated GaN *p*-cap (the highly Mg-doped layer between the active region and the anode contact) of a forward-biased InGaN/GaN LED. At least two distinct peaks were observed in the EDCs. The lower-energy peak was attributed to photoemitted electrons excited in the band bending region (BBR, near the surface of the GaN *p*-cap) by the light generated in the active region. The higher-energy peak was ascribed to the presence, at the *p*-cap surface, of a population thermalized at the bottom of an upper valley of the conduction band (CB) of GaN, derived from high-energy electrons excited by Auger recombination processes in the active region.

These observations of Auger signatures in optically and electrically pumped III-nitride light emitters could be the denouement of the droop debate were it not for the experimental detection limits: a 10^{-6} ratio between detected Auger current and current lost due to droop was estimated in Ref. 12, while the contribution of Auger recombination to all lost charge carriers was determined to be at least 1% in Ref. 13. Yet, these small figures notwithstanding, the detection of Auger-excited electrons was considered evidence that Auger recombination plays a dominant role in LED droop. As high-energy carriers excited by Auger recombination have to travel a distance before emerging from the cesiated surface or being captured by UV wells, a critical question concerning the quantitative interpretation of both experiments arises: How quickly hot electrons decay towards the bottom of the CB while diffusing away from the QWs where Auger processes take place?

A first attempt to answer this question was provided by a recent analytic-band Monte Carlo study of carrier transport in GaN-based LEDs,[17] showing that a sizable fraction of Auger-excited electrons trapped in conduction band side-valleys may reach the *p*-contact without relaxing to the band edge. However, the structure simulated in Ref. 17 is dissimilar from the one investigated experimentally, e.g., the electron blocking layer (EBL) is not included and the number, composition, and thickness of the QWs are different. From a numerical standpoint, Auger and other recombination processes are modeled in Ref. 17 as scattering mechanisms, but self-consistency may not be achieved in a Monte Carlo framework treating on the same footing microscopic processes with so different time scales such as interband recombination and intraband carrier dynamics. Convergence issues aside, since the recombination scattering rates have been derived in Ref. 17 from an empirical *ABC* model, the

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conclusions are, of course, a direct result of the selected *ABC* coefficients. More critically, the simulation of Auger-induced leakage is beyond the *predictive* capabilities of analytic-band Monte Carlo approaches because the excited carriers are promoted to regions of the Brillouin zone, where the electronic structure cannot be approximated in terms of valleys, see Fig. 1. (Discussions of the limits of classical analytic-band Monte Carlo transport simulation can be found, e.g., in Refs. 18–20.)

A quantum transport approach based on a full Brillouin zone (BZ) description of the electronic structure being not available yet, one may assume that Auger-excited electrons behave almost as bulk electrons before relaxing to the band edge. Within a full-band Monte Carlo (FBMC) framework,[21] we will discuss two key assumptions implied in the description proposed in Ref. 12: (i) the presence of a satellite valley in the CB (labeled *L valley* in Ref. 12) at an energy of 0.9 eV above Γ_1^c, compatible with the observed higher-energy peak and (ii) a relaxation rate from that satellite valley to Γ_1^c slow enough to allow an important fraction of *L valley* electrons to reach the BBR before decaying to the bottom of the CB.

As for the first assumption, the authors of Ref. 12, in commenting the state of the art concerning the electronic properties of GaN, claimed that no consensus was established yet concerning the position of its satellite valleys, reporting indirect measurements scattered between 0.29 and 1.49 eV (Refs. 22–26) in contrast with higher theoretical values around 2.0 eV predicted by first-principles calculations.[4,27] Motivated by this controversy, they presented in Ref. 28 a photoemission spectroscopy study of *p*-doped GaN activated to negative electron affinity, claiming the first direct experimental determination of the energy of the satellite valleys in wurtzite GaN. The estimated value of 0.9 eV reported in Refs. 28 and 29 stands out in disagreement with all modern density functional theory (DFT) electronic structure calculations,[4,20,27,30,31] which place satellite valleys in GaN slightly above 2 eV. The question is, of course, if we are willing to dismiss DFT calculations as a reliable tool for the determination of the electronic structure of III-nitrides.

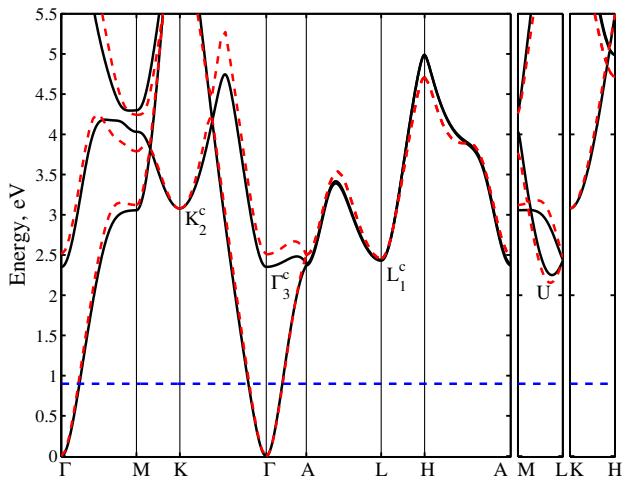

FIG. 1. Conduction band of GaN computed with NL-EPM (black solid lines) and DFT-HSE (red dashed lines). A blue dashed-dotted line marks the energy of the hypothetical satellite valleys suggested by electron emission spectroscopy (EES) experiments.[28,29]

In order to place the question fairly before the reader, it should be mentioned that these first-principles studies are validated by a large body of experimental work on the high-energy critical points of GaN (van Hove singularities) measured by spectroscopic ellipsometry.[16,32–38] Ellipsometric measurements provide a reliable estimate of 2.6 eV for the Γ-*L* energy separation, as fully confirmed by the quasiparticle electronic structure calculations reported by de Carvalho *et al.*[39] when excitonic effects are included by solving the Bethe-Salpeter equation.[40] If we were to question the assignment of the energy transitions to the critical points of the dielectric functions in GaN, we should also reconsider optical studies of AlN[35] and AlGaN alloys[38] that demonstrate the continuous change of the higher energy features. Studies of highly doped GaN did not provide any indication of renormalization effects for transitions related to high energy critical points, let alone low satellite valleys in the conduction band.[37] It is difficult to obtain an electronic structure of GaN having satellite valleys at 0.9 eV (corresponding to the green dashed-dotted line in Fig. 1) with a semi-empirical full-Brillouin-zone approach. If such a small energy difference is imposed, a strong anisotropy and non-parabolicity of the Γ valley should be observed toward the *L* point,[40] in disagreement not only with experiments indicating an isotropic and almost parabolic mass up to 0.6 eV (Ref. 36) but also with well-assessed low-field mobility data.[41,42]

On the other hand, differing experimental assessments of the position of the satellite valleys in GaN were based on indirect measurements,[22–25] and other reports[26] are probably not indicative of bulk properties. More critically, a complete understanding of the complex nature of the GaN cesiated surface is still lacking, since the electron affinity is not likely to be the only electronic property affected by the cesium deposition. Since a DFT study of the electronic structure of a GaN polar surface with cesium and dopant atoms is not yet available, we assumed here a perfect crystalline structure, neglecting cesium-related atomistic effects. Fig. 1 shows the details of the CB in GaN, as computed with the nonlocal empirical pseudopotential method (NL-EPM)[43,44] and with density functional theory based on hybrid functionals (DFT-HSE)[45,46] as implemented in the VASP code.[47,48] (The Hartree-Fock contribution to the hybrid functional was 28%.) These results are in good agreement with each other and with state-of-the-art *ab initio* calculations.[4,27,31] According to the NL-EPM bands, which will be used in the following transport analysis, the lowest secondary valley in the CB (labeled U in Fig. 1) lies along the L–M segment at 2.25 eV above Γ_1^c, while L_1^c is a saddle point, and higher local minima are at 2.35 eV (Γ_3^c) and 3.1 eV (K_2^c) above the CB minimum. The NL-EPM density of states (DOS) of the CB is reported in Fig. 1(b) of Ref. 11, where the peaks corresponding to the satellite valleys are clearly visible.

The second assumption in Ref. 12 is that electrons collected in the satellite *L valley* would not undergo significant relaxation towards Γ_1^c before reaching the BBR, so that the *L valley* would act as a source of high-energy electrons for emission into the vacuum. The presence of long-lived upper valleys with a scattering time to the bottom of the Γ valley of about 1 ps was estimated by Wu and collaborators,[25] by fitting experimental data obtained with a two-color

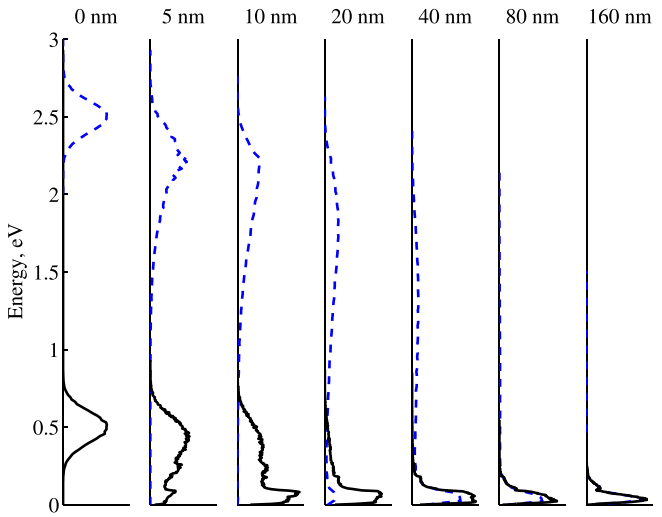

FIG. 2. Kinetic energy distribution of electrons leaking just above the EBL (black solid lines) and excited by Auger recombination (blue dashed lines) in the GaN *p*-cap, as simulated with FBMC, for an applied bias $V_a = 3$ V at increasing distance from the interface with the EBL. Within the first 20 nm, most Auger-excited carriers still populate the satellite valleys located in K_2^c, Γ_3^c, and along the L–M segment (see Fig. 1 and the corresponding density of states reported in Fig. 1(b) of Ref. 11, where the peaks corresponding to the satellite valleys are clearly visible), whereas at a distance comparable with the EBL thickness their distribution is already dominated by carriers in the Γ_1^c valley at the bottom of the CB. At 160 nm, from the EBL, no carriers (out of 10^5 simulated) were found in the upper valleys.

femtosecond pump-probe technique with a rate equation model governing the dynamics of the hot-electron system. The authors of Ref. 12 estimated that high-energy Auger electrons, quickly thermalized in the *L valley*, cross the GaN *p*-cap with a transit time estimated at 1 ps, corresponding to a velocity of 2×10^7 cm/s. However, such a velocity implies ballistic transport, which seems inconsistent with the premise that *thermalized* electrons populate the satellite valleys. Rate equation models are probably not appropriate to describe intervalley transitions, but, even adopting such an approximation, a simple calculation leads to results in disagreement with the interpretation proposed in Ref. 12. As diffusion is the only viable transport mechanism in a region where the electric field is negligible, in order to have a sizable fraction of carriers thermalized in satellite valleys with a mobility of, say, 100 $cm^2V^{-1}s^{-1}$ (but probably lower in the highly doped *p*-type cap) covering distances of the order of 200 nm, one would have to assume a scattering time from *L* to Γ in excess of 100 ps, while intervalley deformation potential scattering times computed, e.g., by Goodnick *et al.*[49] and by our group[21] predict much smaller values (in the range of 0.1 ps). In addition to deformation potential scattering, polar optical emission is also a very efficient relaxation mechanism in GaN, since the LO phonon energy is 90 meV and the coupling is stronger by one order of magnitude compared to other polar III-V materials.[50]

The analytic-band Monte Carlo simulations presented in Ref. 17 seem to be in disagreement both with these basic considerations and with the experimental results they are supposed to validate. In fact, the peak related to upper-valley electrons in the calculated distribution function at the *p*-contact is about four orders of magnitude below the Γ valley peak at the highest injection condition considered (400 A/cm^2, see Fig. 2(b) of Ref. 17, on logarithmic scale), which is probably too large an estimate according to our previous considerations, yet too small with respect to the experimental EDCs, where the higher-energy peak is already visible on a linear scale at much lower current densities (see Fig. 2 of Ref. 12). In an analytic-band descriptions of carrier transport,[17,51–53] the electron population in the satellite valleys depends on several critical quantities: (i) the effective mass tensors and nonparabolicity coefficients, which are intrinsically ill-defined because of the shallowness of the satellite valleys and (ii) the intervalley phonon energy and deformation potentials, which may be regarded as fitting parameters in a simplified treatment of carrier-phonon coupling.

In order to overcome these limitations, we have investigated hot electron transport in the GaN *p*-cap using a FBMC simulator[21] that incorporates the full details of the phonon dispersion and the NL-EPM bands reported above.[54] FBMC provides a rigorous full-Brillouin-zone treatment of deformation potential scattering at high energies, which represents the most critical feature for the present study. The accuracy of FBMC has been demonstrated, e.g., in predicting the multiplication gain and noise properties of GaN-based avalanche photodiodes (APDs)[55] and the nonstationary transport effects observed by time-resolved electroabsorption measurements.[56] Transport of hot electrons through the GaN *p*-cap was studied by simulating the injection, at the heterointerface between the EBL and the *p*-cap, of two populations representing electrons leaking just above the barrier and highly energetic Auger-excited electrons, respectively. Fig. 2 reports the simulated EDCs of the two populations in the GaN *p*-cap at increasing distance from the interface with the EBL. It may be observed that both populations thermalize at the bottom of the CB in less than half the *p*-cap thickness, losing memory of their origin before reaching the BBR.[57]

Our calculations were performed assuming a low electric field (100 V/cm) in the *p*-doped region, in the limit of diffusive transport. A small fraction of electrons in the upper valleys ($\approx 10^{-3}$) may cover distances of the order of 200 nm if an electric field of the order of 1 kV/cm drifts them towards the *p*-cap surface. Higher electric fields, which could develop in the *p*-doped region under very high injection conditions, do not seem consistent with the Mg concentration ($\approx 1.8 \times 10^{20}$ cm^{-2}) and the maximum current density (≈ 100 A cm^{-2}) reported in Ref. 12, see also Fig. 6(b) of Ref. 58.

In principle, carrier distributions should be obtained from a complete device analysis, accounting for quantum effects, vertical carrier transport, Auger, defect-assisted and radiative transitions within a full-band framework. Such a model is not available yet (quantum effects were ignored in Ref. 17 and recombination processes were still obtained from *ABC* models, as in semiclassical drift-diffusion approaches), nor necessary to support our conclusions, as hot electrons injected beyond the EBL (with energies compatible with Auger final states) relax before reaching the surface whatever their origin and initial distribution is, carrier dynamics being fast enough to drive the electron population back to equilibrium. We also performed analytic-band simulations to study the sensitivity of the results with respect to intervalley energies and coupling constants (a full-band implementation is not well suited to this task, since, as

already noted, these quantities cannot be changed independently). Adopting the material parameters in Ref. 59, and launching all carriers in the upper valleys (a very conservative choice), we found that, regardless of valley energies, we need to unrealistically decrease the intervalley scattering rate by at least two orders of magnitude in order to observe a sizable fraction of hot electrons near the surface.

A critical aspect of the experiment which would require further investigation is the spectral shift of the EDCs with increasing applied bias.[29] The effect was attributed by the authors of the experiment to an increasing fraction of the forward bias dropping across the BBR, while the electric field would remain negligible in the bulk of the *p*-cap. As a result, the Fermi level at the surface of the semiconductor would be lowered with respect to the bulk value in the *p*-cap, leading to an apparent high energy shift of the electron distributions. Therefore, the hot electrons observed in the experiment are not, after all, so hot as they might seem. In fact, at the lowest current (3 mA), the higher-energy peak is located just 0.4 eV above the bulk conduction band edge (see Fig. 2(a) of Ref. 29), which, considering the experimental uncertainties, is not far from the electron distribution that one would expect assuming a thermalized population in the Γ valley tunneling across the BBR. Moreover, if a localized bias-dependent voltage drop exists, it is not likely to be uniform across the 10 μm-wide cesiated holes surrounded by the Pt electrode.[60] A more accurate simulation of the experiment would require additional details about the complex device under study, in particular, at the *p*-cap surface, e.g., the Mg doping profile should be investigated to assess possible surface segregation.[61] A detailed analysis of the experimental $I(V)$ characteristics, such as that performed in Ref. 58, would be essential to better understand not only the effects of cesiation but also the possible role of drift-leakage mechanisms, as electrons leaking above the EBL by either Auger recombination or quasi-ballistic transport may drift away from the active region in high-injection conditions.[9,58,62,63]

The experimental uncertainties discussed above are not detrimental to the conclusions of the present work, namely, that the interpretation provided in Refs. 12 and 29 is not consistent with the current theoretical understanding of carrier transport in GaN. We do not imply that Auger recombination and Auger-induced leakage[9] play a negligible role in LED droop, but that an Auger signature can hardly be recovered from the experiment presented in Ref. 12, the measured EDCs being probably uncorrelated with the carrier distribution in the active region.

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