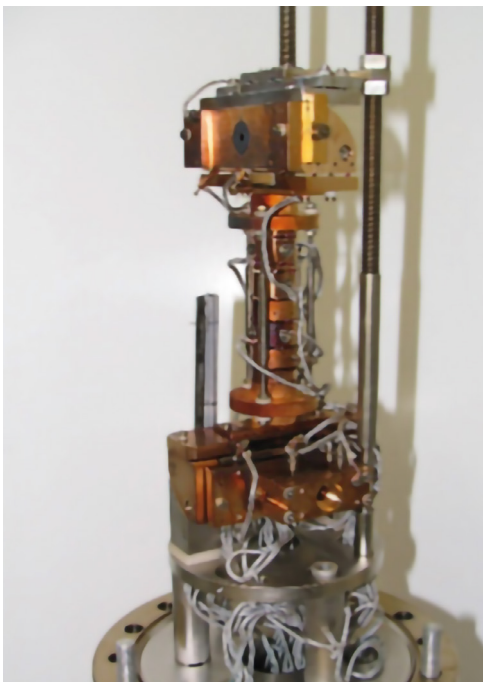


HOW ELECTRON EMISSION UNCOVERED THE LAST MYSTERY OF NITRIDE LEDS



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The remarkable energy efficiency of GaN-based white LEDs has made them the cornerstone of modern lighting. Yet behind this global commercial triumph lay a lingering mystery: why do these LEDs lose efficiency at high currents? That question has finally been answered, thanks to a groundbreaking physics experiment based on electron emission.

<https://doi.org/10.1051/photon/202613724>

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Historical context

Sometimes, technological breakthroughs with major societal impact emerge ahead of our fundamental scientific understanding. The rise of LED lighting is a striking example: high-performance LEDs reached the market even as key aspects of the physics behind GaN, the material at their core, remained poorly understood. In fact, the 2014 Nobel Prize in Physics was awarded to three materials scientists not only for their technical achievement but also for the profound societal impact of their work. While science initially struggled to explain how

these imperfect materials could emit light so efficiently, it has now caught up, uncovering why those same LEDs lose efficiency at high currents.

Among the key factors behind the success of nitride LEDs (GaN and its InGaN and AlGaN alloys) were major advances in material science, notably the improvement of GaN crystal quality through a two-step growth process on sapphire substrates, and above all, the achievement of p-type doping via the activation of magnesium acceptor dopant atoms. This activation, first discovered accidentally under electron irradiation and

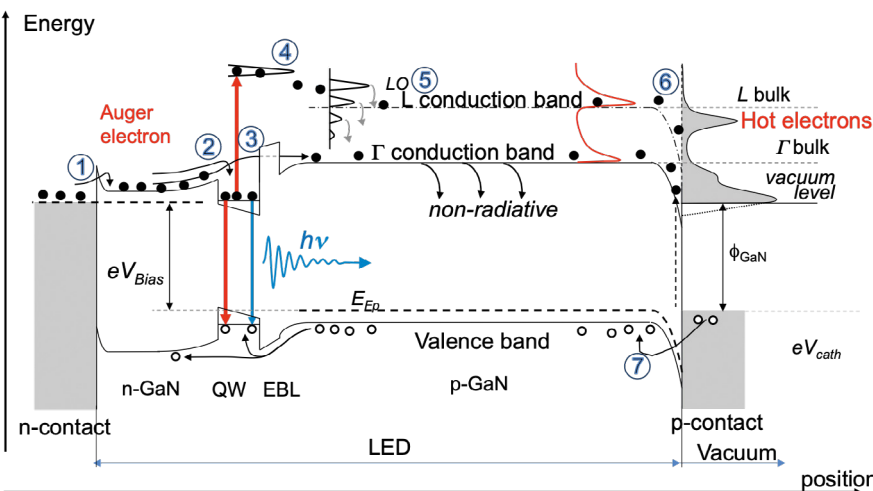
later optimized through thermal annealing, ultimately led to the Nobel Prize together with the discovery of the nucleation layer which leads to materials with high crystalline order. However, the level of p-type doping remained limited compared to n-type doping which might lead to holes injected from the p-side (⑦ in Fig.1) being fewer than electrons injected from the n-side (① in Fig.1). This imbalance might occur in the central region of the LED, composed of InGaN quantum wells, where electron-hole recombination takes place to generate light (② in Fig.1). The non-recombining excess electrons

might leak through the active region into the p-doped layer where they recombine non-radiatively, failing to produce photons. This electron leakage from the active quantum wells (shown as ③ in fig.1) translates directly into a loss of luminous efficiency. The solution came in the form of a quantum barrier, an electron blocking layer (EBL), made of the large bandgap AlGaIn alloy, inserted between the quantum wells and the p-region to block electron overflow. Thanks to this quantum engineering, nitride LEDs can now reach internal quantum efficiencies of up to 90%, meaning that 90% of the injected electrons are converted into photons. These devices are at the heart of the lighting systems found today in our homes, streets, TVs and cars. The LED emission, that use a transition between two energy levels (conduction band and valence band), is by nature monochromatic, at a wavelength that can be adjusted by the Indium content in the InGaIn quantum well. With about 20% In, the LEDs are emitting in the blue. To get white light, one covers the chip with phosphors which absorb part of the blue emission and reemit yellow light, both

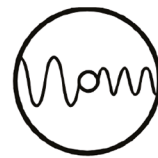
blue and yellow resulting in white light. The story could have ended there, with a global commercial success and lighting products boasting luminous efficiencies around 150 lm/W, far surpassing competing technologies to the point where competition virtually disappeared. But there was still a small flaw in this triumph: when driven at high current, these LEDs lose efficiency. At high power, their performance can drop by a factor of two or three, a phenomenon known as *efficiency droop*.

In the 2010s, this issue began to stir intense debate within the academic community. Guided by the known challenges of p-type doping and electron leakage, many researchers believed the problem once again lay in these areas: under high injection, the proportion of electrons crossing the active region and even the electron blocking layer (EBL) was thought to increase (③ in Fig.1). To counter this detrimental effect, various structural modifications were attempted, such as adjusting the height of the EBL, changing the number of quantum wells, or altering doping profiles. These studies, often carried out in low ●●●

Figure 1. LED band profile and schematic illustrating the Auger effect, which can populate the higher valleys, and electron emission into the vacuum through a cesiated surface. Various mechanisms at work in an LED are illustrated, respectively electron ① and hole ② injection, electron capture in the light-emitting quantum well ②, electron overflow from the active quantum well ③, Auger non-radiative recombination generating hot electrons ④, hot electron energy relaxation into Γ and L bands ⑤, surface energy relaxation generating the electron energy distributions observed outside the semiconductor ⑥.



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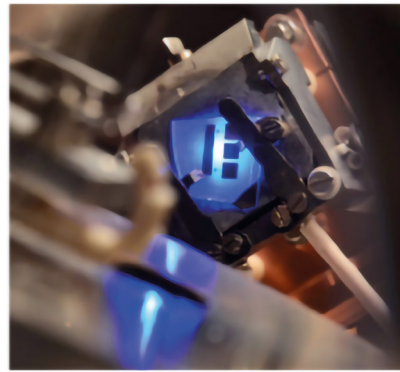
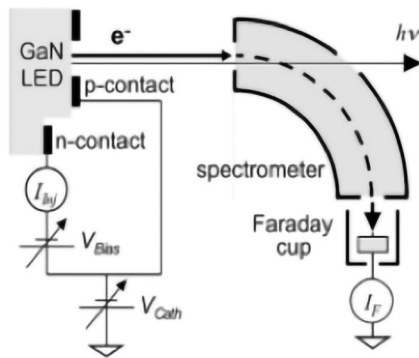


Figure 2. Experimental set up with LED and electron analyzer. Blue LED in operation during measurement, emitting both blue photons and electrons. (photo credit: Ph. Lavalie, École Polytechnique)

efficiency LEDs, met with ambiguous results as they did not improve over state-of-the-art LEDs, suggesting that the root cause had likely not been identified. An alternative and not entirely incompatible explanation also existed: the Auger effect (recently renamed the Auger–Meitner effect). During the recombination of an electron with a hole in an InGaN quantum well, the released energy can, instead of generating a photon, be transferred to another electron (or hole), giving it a large kinetic energy within its band (④ in fig.1). This excess energy is then dissipated as heat (phonons) (⑤ in fig.1). This process directly reduces luminous efficiency as an electron hole pair has disappeared in the process and becomes significant when carrier densities are high (its probability scales roughly with the cube of the charge density n^3) and thus under high current injection. This led to the emergence of the so-called *ABC models*, which describe LED efficiency as a function of current, incorporating contributions from defect-related recombination ($A \cdot n$), radiative recombination ($B \cdot n^2$), and Auger recombination ($C \cdot n^3$). These models provided remarkably good fits to experimental data. However, the somewhat arbitrary choice of the A, B, and C coefficients (and later refinements to the model [1]) meant that the argument was not yet watertight. Moreover, proponents

of the electron leakage theory claimed that leakage could itself be modeled with an n^3 dependence. Well known in small-bandgap materials, the Auger process was long considered negligible in wide bandgap materials like nitrides, where energy and momentum conservation make such interactions unlikely. For this reason, the hypothesis initially seemed implausible. In 2007, a precise analysis of the change in carrier lifetime in photoluminescence concluded that an n^3 mechanism, seen as a signature of an Auger effect, explained the data. Then, around 2011, Auger processes were revisited theoretically [2], taking into account various *indirect* mechanisms. Specifically, the involvement of phonons or changes in wavefunction extension caused by defects, alloy disorder, and other factors can relax momentum conservation, making the process 100 to 1000 times more probable than previously predicted. Doubt began to creep into the nitride LED community, exposed to several alternative explanations: could the droop indeed be linked to the Auger effect? What was missing was a direct, irrefutable experimental proof that could reveal the true origin of the droop. And that proof finally came through a decisive physics experiment where the hot electrons generated by an Auger event could be observed.

The decisive physics experiment

Electron emission is, historically, a technique from surface physics seemingly far removed from the internal physical mechanisms invoked to solve the issues surrounding LEDs. It is at the root of the photoelectric effect. Its basic principle is to observe the energy of electrons emitted from a surface when photoexcited as they retain the energy they had inside. To do this, electrons must first be given enough energy to escape the material. This gives a relation between the photon energy and the maximum energy of the outgoing electron which led Einstein to the photon hypothesis and Millikan to determine the value of the Planck constant by relating the maximum energy of the photoemitted electron from metals to the photon energy. The principle of the experiment carried out in LEDs is to use emitted electrons to measure electron energies *inside* the semiconductor. To study semiconductors, it is beneficial to lower the vacuum level, by coating the surface with a monolayer of cesium, which reduces that level so much that an electron in the semiconductor conduction band has a higher energy than the one in vacuum. Analyzing the energy of electrons emitted then probes the energy distribution of electrons above the conduction band near the surface. This turns out to be crucial for the identification of the Auger processes occurring inside materials. In the Auger effect, an electron initially thermalized at the bottom of the semiconductor conduction band, the so-called Γ conduction band, gains the energy of the electron-hole pair, the bandgap energy, either within that same band or in higher ones (④). The electron relaxes down its energy by returning to the minimum of the Γ valley, so-called Γ point, or by transferring to another higher lying conduction band (L band, ⑤ in Fig.1). In that latter case, an electron in the L valley can relax part of

its energy toward the bottom of that valley, the L point, but may also transition back to Γ . These intra- and intervalley relaxations occur through the emission of phonons, primarily longitudinal polar-optical (LO) phonons with an energy of 91 meV in GaN. In all cases, if the initial electron energy is sufficient (that is, higher than the energy separation between the Γ and L valleys plus the 91 meV of the LO phonon), electrons will populate both valleys. If the time required for the Auger-created hot electrons to diffuse to the surface is shorter than the complete relaxation of all electrons from L to Γ , then some electrons with the energy characteristic of the L valley can be emitted into vacuum. In practice, things are a bit more complex: the band profiles near the surface are not flat but bent, so the energy of the Γ and L valleys decreases near the surface, and hot electrons can occupy a range of energies between the valley positions deep in the material and those at the surface (⑥ in Fig. 1). To observe such hot electrons, one simply needs to inject electrons and holes into the quantum wells with concentrations large enough to induce the Auger effect. This is done by forward-biasing an LED. As shown in Figure 1, this forward bias allows electrons to be injected from the n-type region into the quantum wells but it does not give them high kinetic energy: they are injected in GaN and InGaN and relax to the Γ minimum of the main conduction band. If they traverse the

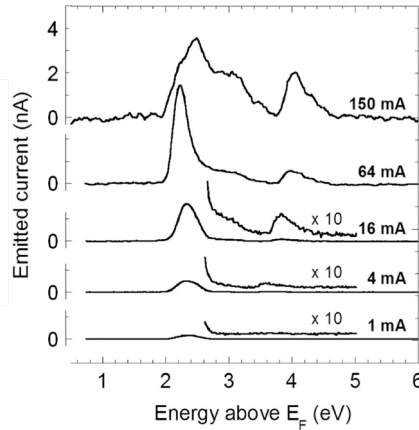


Figure 3. Electron emission spectra for various LED currents showing peaks related to the surface (3), Γ (2) and L (1) valleys.

active region (through leakage), they will be emitted with the energy corresponding to the Γ valley of the p-side. In contrast, if recombination occurs in the InGaN well through the Auger process, electrons will populate the higher-energy valleys accessible to them. A collaboration carried out experiments first at the École Polytechnique in Palaiseau, and later at the University of California, Santa Barbara. The LED, engineered with perforations in its upper contact to allow electrons to pass through, was placed facing an electron spectrometer (shown in the photo at the top of the article) in an ultra-high-vacuum chamber. A single atomic layer of cesium was then deposited on ●●●

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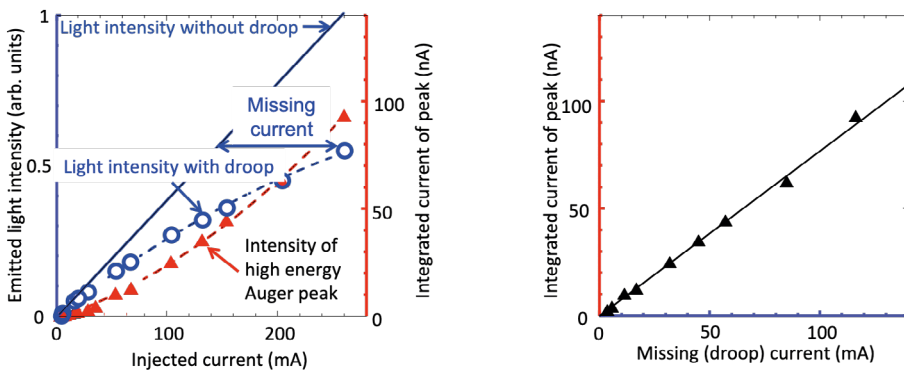


Figure 4. Evolution of the L-valley peak and emitted light intensity as a function of diode current. The extrapolated emission in the absence of droop is shown, defining the “missing current” (left). This missing current is then used to track the intensity of the L-valley peak (right).

the device. After this step, measurements could be performed at room temperature.

Electron energy distribution curves (EDCs) are measured using a cylindrical electron energy analyzer (Figure 2 and top photo). Varying the cathode potential (V_{cath}) allows sweeping the energy. Figure 1 summarizes the energy scale. Note that a hole in the analyser allows taking the electroluminescence emitted by the LED during the electron emission measurement (Figure 2). The electron energy resolution is approximately 50 meV.

The first measurements were carried out on a blue LED composed of eight InGa_N quantum wells containing 18% indium [Ivland]. Taking into account both quantum confinement and internal electric fields in the quantum wells, the transition energy in these wells is 2.75 eV (blue light at 450 nm). The device also includes an Al_{0.15}Ga_{0.85}N EBL, and a 200-nanometer-thick p-doped top layer. When the LED is forward-biased and the current is gradually increased, one, then two, and finally three distinct peaks appear (figure 3). The first, at low energy, corresponds to electrons mainly emitted from the metal excited by the blue light generated by the LED. This first peak extends up to about 0.5 eV, which, when added to the vacuum level, gives an energy of 2.8 eV above the Fermi level, consistent with the photon energy of the emitted light. The second peak, separated from the first through spectral analysis, extends up to around 1.45 eV (estimated at 64 mA) and corresponds to the emission of electrons in the Γ valley. It originates from electrons that have crossed the active region and the EBL as well as from electrons initially in the L valley that thermalized into the Γ valley on their way to the surface. Finally, the third peak, extending up to 2.4 eV, corresponds to a higher-energy valley attributed to the L valley. These “hot” electrons are believed to be a

signature of Auger processes that excite them into the L valley before they diffuse through the p-type GaN to the surface. Their energy distribution broadens in the final, strongly curved region of the band structure. The energy separation between the Γ and L valleys is about 0.95 eV in this case. As the diode’s voltage and current increase, parasitic resistances in the p-contact region cause both the Γ and L valley peaks to shift by roughly the same amount. Tracking these two peaks makes it possible to determine an average separation between the valleys, ultimately estimated at 1 eV. Note that similar electron emission studies were performed on LED structures by optical pumping, *i.e.* photoemission [4]. When the exciting photon energy exceeds 4.2 eV (0.8 eV above the gap), the L-valley signal emerges confirming a value of approximately 1 eV for the Γ -L separation in GaN (and low-indium InGa_N).

Studying how the intensity of the peak associated with the L valley varies with diode current also proves highly informative. At the same time, the emitted light intensity is measured (see Figure 4). Initially, the light output increases linearly with current, but then grows more slowly, with a sublinear behavior that reflects the “efficiency droop.” By extrapolating the initial linear region, one can estimate the emission that would be obtained if this droop did not occur. Comparing the two curves, with and without the droop, makes it possible to determine the current

that did not give emission. This “missing current” corresponds to the portion of the injected current feeding the droop process, *i.e.* Auger recombination. When the intensity of the L-valley peak is plotted as a function of this missing current, a perfect linear relationship appears. In other words, the Auger effect, revealed through the presence of electrons in the L valley, is directly responsible for the loss of quantum efficiency in high-current nitride LEDs.

As further proof of the link between the Auger effect in LEDs and hot electrons, the same measurement was repeated on a simple pn junction without quantum wells where both radiative and Auger recombination are reduced due to the absence of carrier confinement. In that case, only the Γ -valley peak was observed [5]. Thus, roughly a decade of electron emission measurements solved the mystery of the efficiency droop. Have they provided a solution? Partially, yes. Although the problem is intrinsic, it can be mitigated by reducing charge densities. Strategies such as ensuring a more uniform current distribution, increasing the number of quantum wells, or widening them are all feasible approaches and several have already been widely implemented. ●

Acknowledgements

The author acknowledges Prof. Claude Weisbuch (École Polytechnique, France and University of California at Santa Barbara, USA).

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