Supplementary Material to the article
“Ultrafast photoluminescence and carrier localization effects in degenerate indium-rich bulk InGaN”

1. Model description of the interband emission and absorption spectra

Fig. S1 shows low-temperature photoluminescence (PL) and absorption spectra of the investigated sample with the $In_{0.6}Ga_{0.4}N$ active layer. The model description of the obtained spectra was performed assuming Thomas-Fermi density of states (“blurred” near the band edges, see expressions (1) and (2) for ρ_e and ρ_h, respectively, in the main text) and *k*-violating (without momentum conservation) interband transitions. Fermi distribution was assumed for strongly degenerate electrons, and a Gaussian was taken as a simple approach to model the distribution function of nonequilibrium holes. In this case, measured absorption spectrum allows one to estimate, through the fitting procedure, the absorption edge energy ($E_g^*+E_F$) and the characteristic energy span of the valence band “tail” W_h. From the PL spectrum, it is possible, in turn, to determine the span of the conduction band “tail” W_e, evaluate “filling” of the conduction band with equilibrium electrons ($E_F–E_g^*$) and the characteristic spectral width W_0 of the settled, “steady-state” hole distribution as they are localized in the local minima of a random band potential. Finally, comparison of the PL and absorption data allows us to determine the nominal band gap E_g^* and to estimate the average hole localization energy E_{loc} (relative to the nominal valence band top).

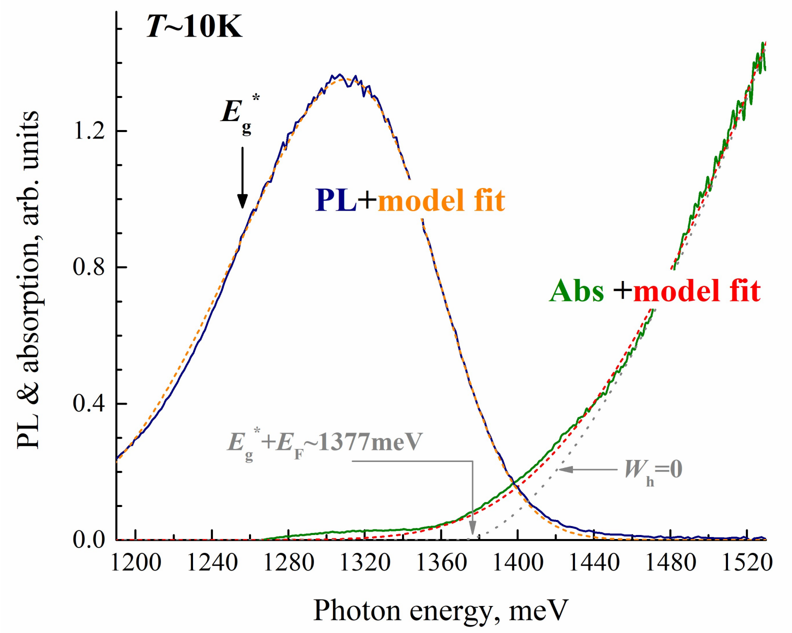

Fig. S1. Low-temperature (*T*~10K) photoluminescence (PL) and absorption (Abs) spectra alongside with the model description of these spectra (dashed lines). The following fitting parameters were used: E_g^*~1262 meV, W_e=53 meV, W_h=43 meV, $E_F=E_g^*+115$ meV, W_0=28 meV and E_{loc}=22 meV. For comparison, the line линия W_h=0 provides calculated absorption spectra for ideal, non-fluctuating bands.

Note that here we analyze the time-integrated PL spectrum, obtained in experiments *without* temporal resolution (irrespective of the excitation regime – either continuous or pulsed, provided that pumping density remains sufficiently weak). Not only localized holes contribute to the measured PL spectrum, but also higher-energy ones that are not yet captured in the potential minima. For this reason, obtained W_0 (and, possibly, E_{loc}) values are somewhat overestimated, in comparison to those determined from the “instantaneous” PL spectra (see Fig. 2*e* and the corresponding discussion in the main text). Nevertheless, the integral contribution of “fast PL” is rather small, and the error in determining W_0 and E_{loc} from the “integrated PL” turns out to be acceptable preliminary estimate for these parameters.

2. Time-resolved photoluminescence: fast and slow response

From Fig. 1*c* in the main text it is evident that the nature of the PL decay changes significantly within the emission spectrum depending on the recording wavelength. Here, Fig. S2 shows the results of fitting the description of the wavelength-dependent PL decay traces by a two-exponential function:

$$PL(\mathrm{t}) = A_{\mathrm{slow}}\times\exp(-t/\tau_{\mathrm{slow}}) + A_{\mathrm{fast}}\times\exp(-t/\tau_{\mathrm{fast}}).$$

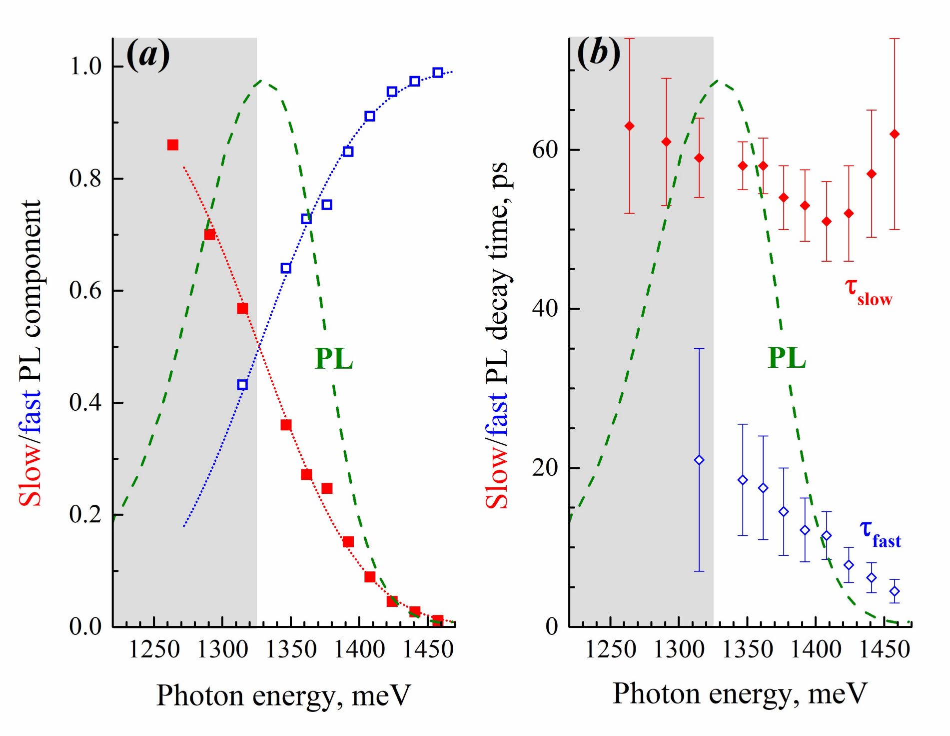

Fig. S2. Time-resolved PL spectroscopy data (symbols): (*a*) relative contributions of the slow ($A_{\mathrm{slow}}/A_{\Sigma}$; $A_{\Sigma}\equiv A_{\mathrm{slow}}+A_{\mathrm{fast}}$) and fast ($A_{\mathrm{fast}}/A_{\Sigma}$) components of the luminescence response, and (*b*) characteristic decay times of these components as a function of the observation wavelength. To facilitate the analysis, the PL spectrum is shown (dotted lines) in both panels; trend lines in panel (*a*) are provided for clarity. The shaded area is the sensitivity limit of the streak camera used, which determines the increase in the error in determining A_{slow} in the long-wavelength region.

In general, slow component of the luminescent response shows decay times of 50-60 ps which does not depend on the detection wavelength. Presumably, it is determined by the *eeh*-type Auger recombination (*CCHC*) – either trap-assisted (see, e.g., [1]), or phonon-assisted [2]. However, available experimental data are insufficient to discuss the question in any further detail.

3. Indirect (*k*-violating) radiative transitions in *n*-InGaN

In the PL analysis, it is assumed (and is generally accepted for III-N structures) that the interband emission is determined by transitions of free electrons to localized hole states, at least if one considers sufficiently low temperatures. In this case, due to the poorly defined momentum of the final state, the *k*-selection rules are relaxed, and not only direct transitions become possible. Here, the simplest qualitative estimates for the applicability of such an approach to the samples under study can be given. Based on the residual electron concentration of $n_{\mathrm{e}}{\sim}8\times10^{18}$ cm^{-3}, the electron momentum at the Fermi level is estimated as $k_{\mathrm{F}}^{(\mathrm{e})} = (3\pi^2 n_{\mathrm{e}})^{1/3} \sim 6\times10^{6}$ cm^{-1}. On the other hand, measured energy scale W_{h} of the valence band “tail” specifies the characteristic spatial extent of localized hole wavefunction and, accordingly, the momentum that can be transferred during interband recombination: $k_p \sim [2m_h W_h/(h/2\pi)^2]^{1/2} \sim 1.1\times10^7$ cm^{-1}, which exceeds $k_F^{(e)}$. The resulting estimate of the characteristic localized holes wavefunction radius (~1 nm) qualitatively agrees with the experimental data (for InN structures with similar electron concentration and, apparently, close structural quality) presented in [3]. Note also that for $n_e \sim 8\times10^{18}$ cm^{-3} the screening radius is estimated at $r_s \sim 2.5$ nm, which means that the screening effects should not affect localized hole states too much. In general, the above estimates allow us to assume that *all electrons* in the conduction band (and not just a fraction of them around k=0) can contribute to the luminescence.

References

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[2] F. Bertazzi, M. Goano, and E. Bellotti. ***Proc. SPIE***, 8619, 86191G (2013)

[3] B. Bansal, A. Kadir, A. Bhattacharya, V.V. Moshchalkov. ***Appl. Phys. Lett.***, 93, 021113 (2008)