Supplementary Information for:

Ultrafast Hot Carrier Dynamics in GaN and its Impact on the Efficiency Droop

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Figure S1: **Phonon mode-resolved electron-phonon scattering rates.** (A) e-ph scattering rates due to each of the 12 phonon modes in a unit cell of GaN at 300 K, shown for electrons and holes with energies within 5 eV of the band edges. Except for mode 12, which is the LO mode, all other modes have similar values of the scattering rate at each energy. The zero of the energy axis is the valence band maximum, and the shaded area is the band gap. For illustration purposes, modes are labeled according to increasing frequency at each q-point, and thus according to their branch index, as color coded in panel (B). (B) Phonon dispersions along high-symmetry lines, showing the phonon branch labeling used in (A).

**Section A. Origin of the stronger average e-ph coupling strength for electrons compared to holes.** As discussed in the main text, the non-polar scattering rate $\Gamma^{(NP)}$ approximately follows the same energy trend as the electronic DOS, $D(E)$, multiplied by an average e-ph coupling strength $\langle g^2 \rangle$, so that $\Gamma^{(NP)}(E) \propto \langle g^2 \rangle D(E)$. The non-polar scattering rate and the DOS are shown in Figure S2A. We find a stronger average e-ph coupling for electrons compared to holes, as seen by the greater ratio of the scattering rate to the DOS for electrons, which is more clearly visible in Figure S2A at excess energies higher than 2 eV. Since the e-ph coupling strength $|g|^2$ is related to the overlap of initial and final states with roughly the same energy, we attribute this difference in the e-ph coupling strength to the different orbital characters in the valence and conduction bands. We quantify the wavefunction overlap by computing the Wannier function (WF) projected DOS (see Methods in main text), shown in Figure S2B, and the spatial spread of the WFs centered on the N and Ga atoms, visualized in Figure S2C. We find that the valence band character is dominated
Figure S2: **Origin of the stronger average e-ph coupling strength for electrons compared to holes.** (A) e-ph scattering rates due to all non-polar phonon modes. The electronic DOS is also shown. Note the higher scattering rate to DOS ratio (and thus the higher average e-ph coupling) in the conduction band. (B) Wannier functions (WFs) projected and total DOS. The projected DOS for WFs centered on N and Ga atoms are shown with blue and red filled curves, respectively. The total DOS, shown in black, is the sum of the two. In panels (A) and (B), the zero of the energy axis is the valence band maximum, and the band gap is shown as a shaded area. (C) Example WFs on N and on Ga are visualized, where the spread Ω of each WF is also indicated.

by WFs centered on N atoms with a spread of $\sim 0.7 \, \text{Å}^2$, while the conduction band character is dominated by WFs centered on Ga atoms with a spread of $\sim 1.7 \, \text{Å}^2$. The fact that the WFs on Ga are more extended than those on N makes them more sensitive to local potential perturbations due to ionic displacements, resulting in the stronger e-ph coupling seen in the conduction band. Note that this difference in spatial extent is related to the polar nature of GaN, suggesting that the stronger short-range e-ph coupling in the conduction band is likely present in other polar compounds.
Figure S3: **Simulated hot carrier dynamics for 0.5 eV initial excess energy.** Holes (left) and electrons (right) are injected with a 0.5 eV excess energy with respect to the band edges, as modeled by creating initial Gaussian carrier distributions with a small energy width. The average carrier concentrations as a function of energy for electrons and holes are shown at various times. The asymmetry between the cooling times of holes (50 fs) and electrons (over 250 fs) is clearly visible, and so are the wiggles in the electron distributions, which are spaced apart by $\hbar \omega_0 \approx 100$ meV, as discussed in the main text.