**Supplementary material**

**Electrostatic contribution to the photo-assisted piezoresponse force microscopy by photo-induced surface charge**

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Fig. S1. Schematic drawing that shows positive surface charges is developed near the sample surface under 405 nm light illumination, inducing an electric field (photo-induced E-field) that is perpendicular to the sample surface (along [001]). The inset shows that such electric field (red arrow) is parallel but opposite to the electric field between AFM tip/cantilever and sample surface introduced by the positive VDC. Take note, our PFM is operating in contact mode, the tip-sample distance here is exaggerated only for the purpose of illustration.

Figure S2 displays the amount of shifting determined at random sample locations over 3 different sample areas. At each sample locations, spectroscopic measurements were performed twice (begin with measurement in the dark followed by measurement under 532 nm light illumination). Since there are no photo-induced surface charges under 532 nm photo-assisted KPFM measurement (not shown), if there are charges injected at the GaN sample surface by the application of VDC, one should observe a constant amount of shifting towards a particular direction when comparing the amplitude curves measured with and without light illumination over all sample locations (right/left-shift along the x-axis, depending on the amount and polarity of the injected charge). In this work, the amount of shifting appear to be random and are fluctuating within the range of about -40 mV to +20 mV. Hence, we conclude that no significant charge injection due to the application of VDC can be observed.

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**Fig. S2.** Amount of shifting determined by comparing the amplitude curves obtained in the dark and under 532 nm light illumination that were measured at 3 different sample areas (represented as Sets 1, 2, and 3) using PFM spectroscopy mode. Within each sample areas (2×2 μm2), spectroscopic measurements were repeated at 8 random sample locations with and without 532 nm light illumination.