Reply to "Comment on 'Observation of Fourier transform limited lines in hexagonal boron nitride'"

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In this Reply, we answer to the comments by Langbein [Phys. Rev. B **100**, 047401 (2019)]. We disagree with the argument that our measured spectral shapes and the extracted linewidths are caused by temporal blinking. We give detailed information on our evaluation process to exclude blinking events. Beyond the question raised in the Comment, we analyze the influence of spectral diffusion. Although spectral diffusion is an ongoing limitation for defect centers in hexagonal boron nitride, we prove that it is not influencing our extracted linewidths.

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We define blinking as a fluorescence change between on and off values, typically caused by charge state instability. We disagree with the statement of the author of the Comment [1] to our recent work that continuous lineshapes in photoluminescence excitation (PLE) scans could originate from blinking. Any blinking would inevitably lead to a discontinuity of the lineshape. To exclude effects of blinking on the spectral lineshapes, we have investigated more than 690 lines. We have inspected each individual lineshape and checked it for continuity. All data with blinking events, examples of which are shown in the original Supplemental Material Fig. 2(a) [2] were excluded from further processing. Blinking would have lead to an underestimation of the linewidth. After the post-processing, 204 lines out of 690 lines have survived the postselection process as shown in Fig. 4(b) in the paper. A faster laser scan velocity in PLE scans results in a higher number of lines surviving the postselection process. We label this quantity with 1-p_{blinking} and plot it in Fig. 1.

A remaining question remains for a scenario where the emitter blinks off and back on much faster than the time of the scan. Such events could lead to an underestimation of the linewidth without discontinuity in the lineshape if some collection bins contain multiple on-off-blinking events leading to an intermediate fluorescence rate. We exclude these events by determining the on- and off-blinking timescale.

In the original Supplemental Material Fig. 1, we determined the characteristic blinking timescale to a value of $t = 0.378$ s. In the same manner, the off-blinking time is determined in Supplemental Material Fig. 1 to be 1.16 s, a factor of 3 times longer than the on-blinking time. So the event of off and back on blinking is a factor of 4 times longer than

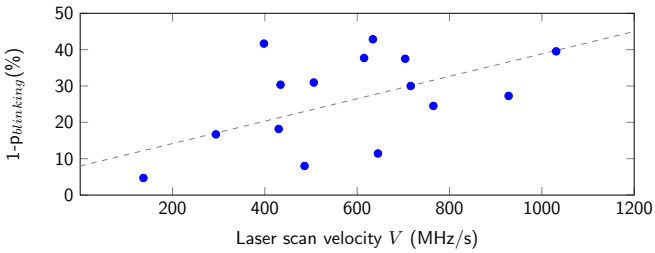

FIG. 1. Likelihood for a PLE scan without effects of blinking plotted against laser scan velocity. The 1-p_{blinking} labels the likelihood of a PLE scan without a blinking event. The 1-p_{blinking} is plotted against laser scan velocity. The graph displays the tendency of increasing 1-p_{blinking} with increasing laser scan velocity. The laser scan velocity is varied from 133 to 1000 MHz/s.

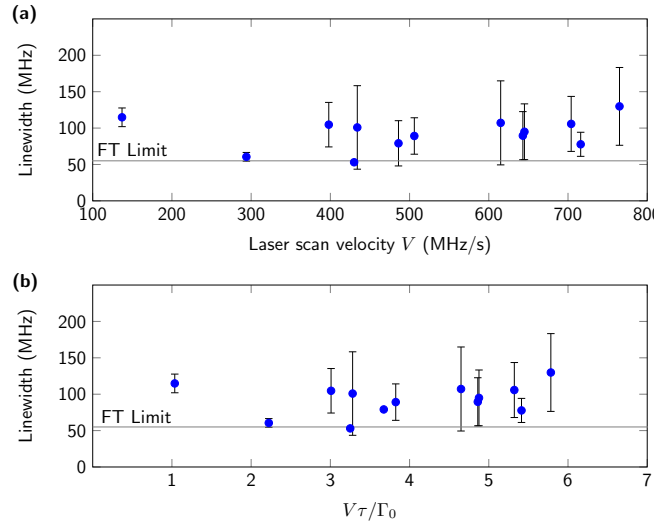

FIG. 2. (a) Mean linewidth for different laser scan velocities at fixed excitation power of 4 μW. The linewidth stays constant at $\approx$ 80–100 MHz over the full range of laser scan velocities. (b) Mean linewidth plotted over normalized laser scan velocity. The laser scan velocity is multiplied with the blinking time constant and divided by the Fourier transform (FT) limited linewidth Γ_0. This value reflects the likelihood to scan over the linewidth Γ_0 without a blinking event for a given laser scan velocity V. A value of 6 means the laser can, on average, scan across the line six times faster than the occurrence of a blinking event. The occurrence of an off and back on blinking during individual bins of the scan is negligible.

the duration of the scan across the full width. So the event of off and on blinking within one bin of the scan is negligible.

However, a more complex question regards ongoing spectral diffusion. Although the postprocessing procedure eliminates blinking as well as large spectral jumps, it does not necessary remove small spectral jitters. Therefore, ongoing spectral diffusion during the PLE line scan could still effect the lineshape. In our measurements, spectral jitters lead to an on average broadened linewidth, which we lay open in Fig. 4(b) of the main paper. Please note that, here, no further postselection is performed on the data. The lineshape changes from a more Lorentzian-like lineshape for the narrow linewidth (as expected for Fourier transform limited lines) to Gaussian lineshapes for the broad linewidth that are broadened by spectral diffusion.

In the experiment, we have varied the laser scan velocity from 133 to 1000 MHz/s in order to test the independence of the inferred linewidth from the laser scan velocity. The fastest laser scan velocity is approximately six times faster than the characteristic on-blinking timescale of 0.378 s. Figure 2(a) shows the average linewidth depending on the laser scan velocity measured with a laser power of 4 μW. The mean linewidth does not significantly change and remains within a range of 80–100 MHz close to the Fourier transform limit. Please note that the mean linewidth is slightly above the Fourier transform limit due to remaining spectral diffusion, whereas individual scans resolve the Fourier transform limit of the homogeneous linewidth. Figure 2(b) recaps the results with laser scan velocity normalized to the blinking timescale.

Please note that we have performed additional checks. We performed PLE scans with different time binnings. Dividing the binning length by a factor 2, therefore, doubling the sampling rate, resulted in the same linewidth. We proceeded with this measurement scheme on a second emitter [main paper Fig. 4(d)] holding a different emission wavelength and a different lifetime as well as a different blinking timescale. Again, we obtained FT-limited lines.

Finally, we note that spectral diffusion is a remaining issue for defect centers in hexagonal boron nitride. The suppression of spectral diffusion will be the focus of future research work. However, the main result in this Reply is to demonstrate that the lines are stable for a long enough time to perform resonant excitation measurements with minor influence from spectral instabilities. This was the purpose of showing the long trace in Fig. 3(b) as well as a blinking one in the Supplemental Material.

[1] W. Langbein, preceding Comment, Phys. Rev. B **100**, 047401 (2019).

[2] A. Dietrich, M. Bürk, E. S. Steiger, L. Antoniuk, T. T. Tran, M. Nguyen, I. Aharonovich, F. Jelezko, and A. Kubanek, Phys. Rev. B **98**, 081414(R) (2018).