Response to the Comment on “Excitons in Molecular Aggregates with Lévy Disorder: Anomalous Localization and Exchange Broadening of Optical Spectra”

In our Letter [1], we predicted exchange broadening and a blue shift of the absorption spectrum with increasing disorder strength $\sigma$ for heavy-tailed disorder with a stability index $\alpha < 1$. We explained these findings by deriving ‘conventional’ scaling laws for the width of the absorption spectrum and the delocalization length. In addition we predicted new features in the density of states and the absorption spectrum, as well as non-universality of the localization length distribution, which we attributed to the appearance of ‘outliers’ in the disorder, i.e. sites with energies outside of the bare exciton band.

The main points of the Comment [2] are that the outliers introduced by heavy tails in the disorder distribution (i) do not lead to deviations from the conventional scaling law for the half width at half maximum (HWHM) of the absorption spectrum and (ii) do not lead to non-universality of the distribution of localization lengths. We show below that the findings reported in our Letter [1] are correct and that the wrong conclusions of the Comment [2] arise from focusing on small $\sigma$ values.

The conventional scaling law for the HWHM ($\equiv$ FWHM/2) in the general case of symmetric $\alpha$-stable Lévy distributions reads

$$\text{HWHM} \sim J(\sigma/J)^{2\alpha/(1+\alpha)}.$$  

(1)

Its derivation was given in our Letter [1] already, generalizing from the special cases of Gaussian ($\alpha = 2$) and Lorentzian ($\alpha = 1$) disorder [3, 4] to arbitrary values of $\alpha$. In the Comment [2], this derivation is repeated without any additional insights (last paragraph of the Comment [2]). In the derivation, segmentation due to outliers (defined by us as sites with deviations from the average energy larger than $2J$, acting as barriers) as well as finite size effects are not taken into account. For $\sigma$ values of interest for many molecular systems ($\sigma \lesssim J$), deviations from the conventional HWHM scaling due to outliers have not been seen for $\alpha = 1$ or 2. However, for $\alpha \approx 0.5$ and smaller clear deviations are visible in this $\sigma$ regime. This is due to the fact that for the same $\sigma$, outliers become more abundant for heavier tails (smaller $\alpha$). By equating $N^*$ and $N_{\text{seg}}$ from Eqs. (3) and (4) of our Letter [1], one can estimate the value of $\sigma$ above which segmentation may be expected to play a significant role (indicated as the shaded region in Fig. 1a).

To demonstrate the deviations from the conventional scaling at smaller $\alpha$ values, we plot in Fig. 1 the HWHM as a function of $\sigma$ for $\alpha = 0.5$ (b) and $\alpha = 0.4$ (c). The red lines are power-law fits imposing the conventional scaling law. The case $\alpha = 0.5$, panel (b), was also given in our Letter [1] but now has many more data points (symbols), also compared to the Comment [2]. For $\sigma \lesssim 0.4J$ the conventional power law fit works well, agreeing with the observation in Ref. [1] and repeated in the Comment; for larger $\sigma$, the conventional scaling clearly breaks down. Moreover, the data points show kinks at which the HWHM suddenly jumps. This is not noise, but may be unambiguously attributed to structure in the high-energy wing of the absorption spectrum which grows in for increasing $\sigma$ as a consequence of segmentation [1]. While both effects are visible already at $\alpha = 0.5$, they become more pronounced for smaller $\alpha$ (panel c). This new data also shows that these features cannot be adequately represented by a power law. Finally, we mention that the straight line in Fig. 2 of our Letter [1] indeed is meaningless without the information how the monomer HWHM scales with $\sigma$; this has no effect on the conclusion of that figure, namely the discovery of exchange broadening.}

Next we demonstrate the non-universality of the localization length distribution due to the segmentation by outliers. While the data used for this purpose in our

![FIG. 1: (a): “phase diagram” showing for which ($\alpha, \sigma$) combinations segmentation is expected to become visible. (b,c): HWHM of the absorption peak as a function of $\sigma$ for (b) $\alpha = 0.5$ and (c) $\alpha = 0.4$. Data points obtained from numerical simulations ($N = 200, 10^7$ realizations), red curve representing a fit corresponding to the conventional localization mechanism (note the deviations from the fit).](image-url)

![FIG. 2: Distribution of normalized localization lengths for small disorder values (a) and intermediate disorder values (b).](image-url)
Letter [1] (Fig. 3 and the $\sigma$-dependence of $\delta N_{\text{loc}}/\bar{N}_{\text{loc}}$) were correct, they were not a good choice, especially not because they were discussed in the context of Ref. [3], where the energy interval in which states are considered was scaled with the width of the absorption band. In Fig. 2, we show that also for an energy interval which is “properly” scaled, the non-universality becomes visible. This figure displays for $\alpha = 0.5$ the distribution of normalized (to the average) localization lengths, obtained using the same scaling of the width of the energy interval as in the Comment [2] (cf. Ref. [4]). In Fig. 2(a) small $\sigma$ values are considered ($\sigma \leq 0.1J$, as in the Comment [2]). As expected from Fig. 1(a) segmentation is not relevant then and the normalized distributions are nearly identical for all $\sigma \leq 0.1J$. Yet, already for $\sigma = 0.1J$ deviations become clearly visible in the appearance of several small sharp peaks at small localization length. Upon increasing $\sigma$ (Fig. 2(b)), these deviations become more and more pronounced and the non-universal character of the distribution is clear [5]. By considering the wavefunctions, the origin of these peaks can be traced back to the segmentation mechanism. We note that also for the fixed energy interval used in our Letter [1], these signatures of segmentation and non-universality can be found.

In conclusion, we have shown that the claims put forth in the Comment are not generally valid, but are instead an artifact of focusing on a regime where segmentation effects are expected to be small. In the Comment it is stated that conventional scaling may break down at high $\sigma$ values, only because $N_{\text{loc}}$ approaches unity. This is a trivial effect and does not explain the additional structure observed in the localization length distributions. As we have demonstrated, the breakdown found by us is more subtle and really is intimately related to outliers.

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[1] A. Eisfeld, S. M. Vlaming, V. A. Malyshev, and J. Knoester, Phys. Rev. Lett. 105 137402 (2010).
[2] A. Werpachowska and A. Olaya-Castro, arXiv:1208.2779v2 [cond-mat.dis-nn]
[3] V. A. Malyshev, J. Lumin. 55, 225 (1993).
[4] S. M. Vlaming, V. A. Malyshev, and J. Knoester, Phys. Rev. B 79, 205121 (2009).
[5] When taking a broader (“correctly” scaled) energy range, the segmentation and the non-universality becomes even more pronounced.