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Resolving ultrafast exciton migration in organic solids at the nanoscale

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The migration of Frenkel excitons, tightly-bound electron-hole pairs, in photosynthesis and in organic semiconducting films is critical to the efficiency of natural and artificial light harvesting. While these materials exhibit a high degree of structural heterogeneity on the nanoscale, traditional measurements of exciton migration lengths are performed on bulk samples. Since both the characteristic length scales of structural heterogeneity and the reported bulk diffusion lengths are smaller than the optical diffraction limit, we adapt far-field super-resolution fluorescence imaging to uncover the correlations between the structural and energetic landscapes that the excitons explore. By combining the ultrafast super-resolved measurements with exciton hopping simulations we furthermore specify the nature (in addition to the extent) of exciton migration as a function of the intrinsic and ensemble chromophore energy scales that determine a spatio-energetic landscape for migration.

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