Review of: "Correlating exciton coherence length, localization, and its optical lineshape"

Josene M. Toldo[1]

1 Aix-Marseille Université

Potential competing interests: No potential competing interests to declare.

Information about the environment of a system can be correlated to lineshapes of spectroscopic transitions. In this paper, the authors present a model to correlate the lineshape of a molecular exciton with exciton localization length at finite temperatures, taking into account static and dynamical disorder. The model is built upon a numerically exact self-consistent treatment, where thermal effects are introduced as excitations about the zero-temperature soliton state and exciton bandwidth and exciton reorganization energy enters as parameters. Using this model, the authors show that, in the limit of a strong localized system, an exciton homogeneous line width is observed to be independent of the temperature, while in the limit of a molecular aggregate, the line width is independent of the temperature.

The connection of lineshapes of molecular excitons and finite-temperature lattice vibrations is an interesting idea. However, the paper is quite technical, and the equations are not easy to follow in many parts, which may limit its accessibility to a broader audience. Some additional context and explanation in certain sections, as well as references to certain equations could improve its accessibility. Yet, the reader would also benefit from more connections with real systems or experiments on which this model is applied. I also would like to add the following remarks/questions:

1. The authors mention specific typical values for the Huang-Rhys parameter and phonon frequency in polymeric systems. Could the authors provide a reference for these values?
2. The authors mention that the model should be valid for a wide range of organic-semiconducting materials. In which situations the model is not expected to work well?
3. Equation 21 determines exciton delocalization length, which may reveal how is the competition between exciton localization (which increases with J) and exciton/phonon coupling (which localizes the state). Is there any proof of the applicability of this expression to a real system? It would be great if the authors could provide an example here.