

UNIVERSAL SCALING OF THE ANISOTROPIC DISPERSION IN 2D EXCITONIC SYSTEMS
AND ITS SPECTROSCOPIC SIGNATURES

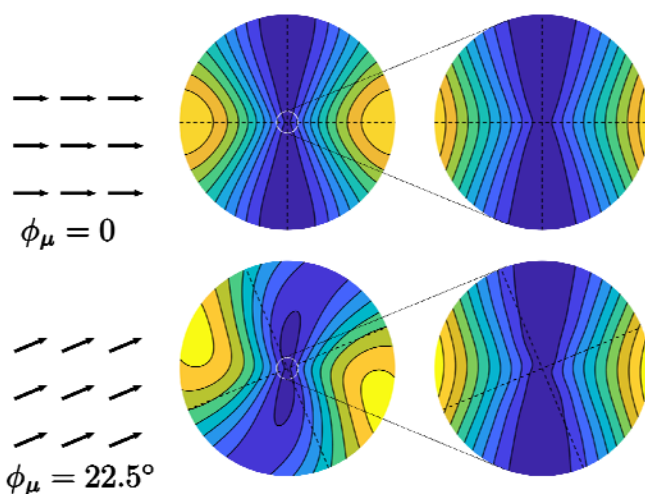
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We derive closed-form expressions for the dispersions in two-dimensional dipolar systems in the small k regime, applicable to a large class of excitonic systems such as molecular aggregates and crystals. For in-plane dipoles, the long-range dipolar contribution leads to a linearly scaling excitonic band along the direction of the dipole near the bright state, while the perpendicular direction is dispersionless up to linear order. We show that such an anisotropic dispersion relation leads to a specific scaling of the system density of states when the bright state is at the bottom of the band. Due to the insensitivity of the long-range coupling to the detailed packing conditions and molecular conformations, these universal scalings allow us to predict numerous spectroscopic signatures in the optical regime. These include absorption linewidths dependent on disorder and temperature, in addition to peak splittings in (transient) absorption spectra. The theoretical predictions of these observables are in quantitative agreement with numerical simulations of molecular aggregates consisting of C8S3 dyes of realistic parameters.



References

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[2] Chuang, C.; Bennett, D. I. G.; Caram, J. R.; Aspuru-Guzik, A.; Bawendi, M. G.; Cao, J. *Chem* **2019**, 5, 1.

Figure 1. 2D dispersion relations of dipolar lattices.