Theory of paired Zhang-Rice excitons in a layered antiferromagnet, $NiPS_3$

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A recent observation of an ultrasharp peak in the photoluminescence (PL) of NiPS3 [1] raises several questions about the nature of its excited states. While the proposal of a many-body exciton—a Frenkel-type local electron-hole excitation transitioning from the Zhang-Rice triplet to singlet (ZRS) state within a zigzag antiferromagnetic (AFM) background—successfully explains both the optical and resonant inelastic x-ray scattering (RIXS) spectra, the ZRS exciton theory still faces many unresolved issues. In this talk, we aim to address several puzzles related to this exotic PL signal by proposing a theory of paired ZRS excitons [2]. Unlike a typical immobile Frenkel exciton, we demonstrate that a single ZRS exciton can propagate along one-dimensional ferromagnetic chains in the zigzag AFM background, and that their pair formation is favored over isolated excitons when two ZRS excitons occupy nearest-neighbor sites along the chain.Three notable puzzles resolved by our new proposal are as follows: First, unlike the isolated dark ZRS exciton, the paired ZRS (PZRS) state has a well-defined entangled optical transition channel for both spin and charge sectors. Second, the small coherent energy scale measured in RIXS for the excitonic state can be attributed to the binding energy between two ZRS excitons. Third, the hypersensitive PL spectra observed under a pressure as low as 0.4 GPa [2] and with minimal Cd doping of less than 1% can be explained by the fragile nature of the excitons [3], which is linked to their characteristic small binding energy.

[1] S. Kang, K. Kim, B. H. Kim, J. Kim *et al.*, Nature **583**, 785 (2020).
[2] K.-X. Zhang, M. Zhang, M. Kim *et al*., submitted (2024).
[3] J. Kim, W. Nat, J. Kim *et al*., Nano Lett. **23**, 10189 (2023).