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Exciton Dynamics in I_2-Intercalated $MoSe_2$ crystals: Temperature-Dependent Luminescence and Kinetic Model

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MoSe₂ layered crystals intercalated with iodine (MoSe₂:I₂) exhibit distinct photoluminescence (PL) due to excitons bound to the halogen molecules, which form neutral isoelectronic centers in the van der Waals gap [1]. At low temperatures, the luminescence is dominated by two sharp zero-phonon lines (ZPLs) labeled A and B at 1.036 and 1.042 eV, respectively, due to exciton recombination bound on iodine [2]. These lines, separated by Δ_{AB} = 5.6 meV are accompanied by a phonon sidebands.

With increasing temperature, a pronounced redistribution of intensity occurs between the A and B spectral components. Initially, the lower-energy A line dominates, but as temperature rises the higher-energy B line grows in intensity at the expense of A. This spectral evolution signifies thermally activated population exchange between the two exciton levels. Notably, the B state (higher energy) has a much faster radiative recombination rate (shorter lifetime) than the A state. Consequently, as thermal excitation promotes excitons from A to B state, the luminescence increasingly originates from the higher level.

Time-resolved measurements show that at low temperatures (T < 30K) the two-level exciton system is not in thermal equilibrium. The decay lifetimes of the A and B emissions differ by nearly a factor of two in this regime, indicating that the decay cannot be described by a single equilibrium lifetime. This nonequilibrium arises because inter-level exciton transfer is slow relative to radiative recombination at low T, owing to the sizable energy barrier Δ_{AB}. A kinetic model incorporating finite inter-level exchange is developed to account for this behaviour. By contrast, in previously studied halogen-intercalated TMDs with smaller Δ_{AB}, exciton populations remain near-equilibrium and a single radiative lifetime suffices to describe the decay. MoSe₂:I₂ thus exemplifies a process requiring an explicit two-level rate-equation model for exciton dynamics.

At higher temperatures (above ~60 K), the bound exciton PL intensity drops rapidly, with an activation energy of ~0.14 eV for this thermal quenching, that is attributed to thermally activated escape of the electrons from the I₂-bound state –an extrinsic self-trapping mechanism [3].

The developed kinetic model quantitatively reproduces the spectral and temporal characteristics of MoSe₂:I₂ exciton luminescence across the 10–150 K range. It captures the temperature-dependent A–B intensity ratio and the distinct A and B decay profiles, including the low-T non-equilibrium and high-T quenching behaviours. Our findings highlight that molecular intercalation can cause unique excitonic dynamics, and demonstrate the importance of including thermally activated population exchange in modeling exciton recombination in such two-level systems.

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