Exciton reactions on carbon nanotubes: an experimental testbed for critical dynamics

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The one-dimensional coalescing random walk \((X + X \rightarrow X)\) is a paradigmatic reaction-diffusion system due to both its exact solvability [1,2] and the experimental observation of nonclassical kinetics at asymptotically long times [3]. The solvability rests on the anticommutative property of intersecting trajectories of particles that react instantly and at short range; however, the validity of these assumptions in real systems has not previously been tested by experiment. We have shown that exciton-exciton recombination (fusion) on carbon nanotubes provides a platform for quantitative studies of critical kinetics in a simple non-equilibrium system [4]. Under high excitation density we observed a crossover in the exciton density \(n\) between regimes of classical \((n \propto t^{-1})\) and anomalous \((n \propto t^{-1/2})\) scale invariance as predicted by renormalization group [5] and approximate [1] theories, arising from a finite reaction probability of \(\approx 0.2\) per encounter. At long times the exciton population per nanotube exponentially approaches unity (i.e. a finite size effect), allowing calibration of the exciton density and hence a demonstration of universality extending over both classical and critical regimes. Under low excitation, the early kinetics followed a Smoluchowski-Noyes form \(dn/dt \propto n^2t^{-1/2}\) rather than the asymptotic \(dn/dt \propto n^3\), providing direct evidence for the spatial self-ordering that precedes critical scale invariance. We studied the re-emergence of microscopic detail at the classical-nonclassical crossover, which is abrupt and nonmonotonic due to competition between temporal and spatial averaging of critical fluctuations (i.e. finite reaction rate and range). It appears that real-world experiments will require more complete descriptions of the interactions than is available in existing models.


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