

# PHASE TRANSITIONS IN ELECTRON SPIN RESONANCE UNDER CONTINUOUS MICROWAVE DRIVING

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The control and detection of magnetization arising from a polarized ensemble of unpaired electron spins forms the basis of electron spin resonance, a powerful spectroscopy tool for studying paramagnetic materials placed in a static external magnetic field. The underpinning key principle for this technique is the application of oscillating magnetic fields close to or at the electronic Larmor frequency (usually in the microwave regime) to generate non-equilibrium distributions of populations and coherences between quantum states that lead to detectable signals. The evolution of systems of isolated or only weakly coupled paramagnetic centres under the effect of these fields is well understood. A more challenging problem is to predict the response of strongly coupled electron ensembles to such perturbations, particularly in samples in the solid state in which anisotropic components of the electronic interactions are not averaged out by thermal motion. Insight into the dynamics of strongly coupled, microwave driven electronic ensembles is also needed in order to improve our understanding of dynamic nuclear polarization (DNP), which is an out-of-equilibrium technique to enhance the sensitivity of nuclear magnetic resonance applications by orders of magnitude — in particular, this concerns the cross effect and thermal mixing DNP mechanisms.

Here we shed light on the non-equilibrium stationary states of a strongly interacting electronic ensemble under continuous microwave driving and subject to dissipation to the environment. We model the dynamics of this system in terms of a Markovian master equation and use a mean-field approximation to compute the steady state phase diagram. This reveals phase transitions between states of high and low electronic polarisation as well as the emergence of a critical point that displays Ising universality. These features are controlled by the distribution of the disordered electronic spin-spin interactions. The uncovered mean-field transitions imply the emergence of metastable

states and accompanying intermittent dynamics, which we confirm numerically through simulations of small systems. Our results suggest that under appropriate conditions collective phenomena such as metastability, phase transitions and critical behaviour should be observable in driven-dissipative, paramagnetic systems. These predictions complement those of conventional theoretical approaches, based, e.g., on the so-called spin-temperature which, due to their restriction to certain parameter regimes, would only predict a homogenous quasi-equilibrium state.