Misjudging frustrations in spin liquids from oversimplified use of Curie-Weiss law

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I. INTRODUCTION

Quantum fluctuations prevent settling down of spins into a classically ordered ground state in magnetic systems even at absolute zero temperature despite strong correlations, giving rise to exciting non-trivial quantum spin-liquid (QSL) state in solids [1]. Such a quantum effect demands strong geometrical frustrations in the lattice, for dimensions higher than 1 to prevent the classical freezing of the spins at low temperature [2–4]. After the prediction of such states by Anderson in ’80s [5], much has been progressed numerically and theoretically to describe the state of spin liquids which have highly entangled quantum phases (e.g. Resonating valence bond solids and liquids) that are spatially separated using non-local topological order parameters and entanglement entropy of ground states [6–9, 11]. Even though, many spin-liquid candidates have been proposed over the last few years [10–16], characterising them experimentally remains obscure till now [17, 18] since the non-local order parameters governing QSLs aren’t associated with conventional Landau ‘symmetry breaking’ during phase transitions [19, 20]. The first and the most popular identifying signature of candidate spin liquids till date, is a large amount of frustration present, indicated by frustration parameter \( f = \theta_{CW}/T_N \) where \( \theta_{CW} \) is the Weiss constant causing deviation from Curie law of susceptibility for paramagnets owing to presence of interactions in the system and \( T_N \) is the Néel temperature or the lowest temperature down to which static moments remain absent in the system. The claims of such exotic states have thus mostly relied on the absence of conventional symmetry breaking into ordered states according to the experimental magnetic susceptibility \( \chi \) and large ground state degeneracy obtained from specific heat measurements. As additional supports, nuclear magnetic resonance and muon spin resonance are used to show presence of local magnetic fluctuations down to the lowest measured temperature [9, 11]. Therefore proper determination of \( \theta_{CW} \) from \( \chi (T) \) curves becomes the factor of utmost importance.

Now, Curie-Weiss (CW) law which had been developed from mean field approximations assuming presence of an effective internal molecular field on a spin due to surrounding spins considering only nearest neighbour (local) interactions that leads to ‘ordering’ in a system during a phase transition quantified by the Landau order parameter (magnetization in the case of a magnetic ordering), obviously fails to describe the ground states of spin liquids which feature non-local topological orders [6]. Nevertheless, it can still be used to estimate exchange interactions \( J^{ex} \) in the system since \( \theta_{CW} \propto \sum z J^{ex} \) from mean field theory [21], where \( z \) are the nearest neighbours. This procedure becomes accurate provided one does a fitting of the magnetic susceptibility with Curie-Weiss law \( (\chi = \frac{C}{T - \theta_{CW}}) \), where \( C \) is the Curie constant related to the effective moment) at sufficiently high temperatures (preferably \( T > 2T_N \)) to avoid criticality near transition temperature as an effect of growing correlation length and order parameter fluctuations [22, 23]. Thus if one fits the susceptibility to mean field form at a temperature where other terms are important then the \( \theta_{CW} \) will vary. Such odd variation in \( \theta_{CW} \) for different temperature ranges are not new and have been seen for many systems having weak/competing interactions on frustrated or magnetic sub-lattices [24–27].
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