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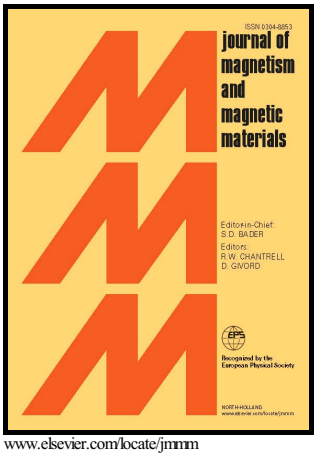
Abhishek Nag, Sugata Ray



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ACCEPTED MANUSCRIPT

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

Abhishek Nag*
Department of Materials Science, Indian Association for the Cultivation of Science, Jadavpur, Kolkata 700032, India

Sugata Ray
Department of Materials Science, Indian Association for the Cultivation of Science, Jadavpur, Kolkata 700032, India and Centre for Advanced Materials, Indian Association for the Cultivation of Science, Jadavpur, Kolkata 700032, India

Absence of a single smoking-gun experiment to identify a quantum spin liquid, has kept their characterisation difficult till date. Featureless *dc* magnetic susceptibility and large antiferromagnetic frustration are always considered as the essential pointers to these systems. However, we show that the amount of frustration estimated by using generalised Curie-Weiss law on these susceptibility data are prone to errors and thus should be dealt with caution. We measure and analyse susceptibility data of $Ba_3ZnIr_2O_9$, a spin orbital liquid candidate and Gd_2O_3, a 1.5 K antiferromagnet and show the distinguishing features between them. A continuous and significant change in Curie and Weiss constants is seen to take place in $Ba_3ZnIr_2O_9$ and other reported spin liquids with the change in the range of fitting temperatures showing the need of a temperature 'range-of-fit' analysis before commenting on the Weiss constants of spin liquids. The variation observed is similar to fluctuations among topological sectors persisting over a range of temperature in spin-ice candidates. On the other hand, even though we find correlations to exist at even 100 times the ordering temperature in Gd_2O_3, no such fluctuation is observed which may be used as an additional distinguishing signature of spin liquids over similarly featureless correlated paramagnets.

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 $f = |\theta_{\rm CW}|/T_{\rm N}$ where $\theta_{\rm CW}$ is the Weiss constant causing deviation from Curie law of susceptibility for paramagnets owing to presence of interactions in the system and $T_{\rm N}$ 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 χ 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 $\theta_{\rm CW}$ from χ (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 parameters (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 $J^{\rm ex}$ in the system since $\theta_{\rm CW} \propto \sum zJ^{\rm ex}$ 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 ($\chi = \frac{\rm C}{T-\theta_{\rm CW}}$), where C is the Curie constant related to the effective moment) at sufficiently high temperatures (preferably $T > 2T_{\rm N}$) 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 $\theta_{\rm CW}$ will vary. Such odd variation in $\theta_{\rm CW}$ 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].

* Email: msan@iacs.res.in

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