From RVB to supersolidity: the saga of the Ising-Heisenberg model on the triangular lattice

1. **Continuum excitations in a spin-supersolid on a triangular lattice**
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   https://doi.org/10.48550/arXiv.2401.16581

2. **Phase Diagram and Spectroscopic Evidence of Supersolids in Quantum Ising Magnet $K_2Co(SeO_3)_2$**
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Recommended with a Commentary by Frédéric Mila, Ecole Polytechnique Fédérale de Lausanne, Switzerland

In 1973, Anderson wrote a famous paper in which he suggested that the ground state of the spin-1/2 Heisenberg model could, under certain circumstances, be a resonating valence bond (RVB) state, i.e. a superposition of states in which singlets are formed on the dimers of dimer coverings of the lattice [1]. One year later, Fazekas and Anderson [2] argued that this must be the case for the anisotropic Ising-Heisenberg spin-1/2 triangular lattice antiferromagnet defined by the Hamiltonian

$$\mathcal{H} = J\sum_{\langle i,j\rangle}[\Delta(S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z],$$

at least in the limit $\Delta \to 0$. When $\Delta = 0$, the model reduces to the Ising model. Since the ground state of this model is infinitely degenerate [3], the energy gain when switching on the transverse exchange must be linear in Δ. Now the energy gain of the ordered state is only quadratic in Δ. So, in the limit of small Δ, the ground state cannot be the ordered state.

Unfortunately this argument was soon proven to be inconclusive [4]: When zero point fluctuations are included, the energy gain of the ordered state is also linear in Δ.

The problem of ordering for $\Delta < 1$ is actually a subtle one. In 1985, Miyashita and Kawamura [5] showed that the classical ground state is infinitely degenerate, and the ordered state is only selected by zero point fluctuations. So one can expect a soft spectrum and strong quantum fluctuations. Still there is no reason to exclude long-range order on the basis of a semi-classical analysis [6]. On the numerical side, the problem with $\Delta > 0$ suffers from a minus sign problem for Quantum Monte Carlo, but when restricted to the ground state manifold of the Ising model, which can be expected to be valid for small Δ, there is a canonical transformation that changes the sign of Δ [7, 11]. So the two models can be expected to have similar properties for small Δ, and the model for $\Delta < 0$ has been shown to have long-range order with Quantum Monte Carlo [8].

To summarize, it is by now generally well accepted that the ground state has a three-sublattice order with one spin along z and two spins with angles θ and $-\theta$ away from that spin, with θ growing from $\pi/2$ to $2\pi/3$ when Δ grows from 0 to 1. This state breaks the translation symmetry in real space and the rotational symmetry around z in spin space, making it a *spin supersolid*, a concept borrowed from the physics of bosons, in which case it refers to the simultaneous presence of a density wave that breaks the translational symmetry and of superfluidity. The only theoretical piece of information that does not fully fit into this picture is the temperature dependence of the Wilson ratio $R = 4\pi^2 T\chi_0/3s$, where χ_0 is the susceptibility and s the entropy. At small Δ, it drops very abruptly at low temperature, a behaviour similar to that of the spin-1/2 kagome antiferromagnet, a system widely believed to be a quantum spin liquid [10].

In the presence of a magnetic field along z, i.e. of an extra term $-h\sum_i S_i^z$ in the Hamiltonian, it is useful to start from the Ising limit $\Delta = 0$ to discuss the physics. In zero field, the ground state manifold consists of all configurations with two spins up and one spin down or two spins down and one spin up per triangle. The magnetic field immediately selects configurations with two spins up and one spin down, leading to three-sublattice order with all spins down on one sublattice and all spins up on the other two. At $h = 6J$, there is a first-order transition into the fully polarized state. For $0 < h < 6J$, there is a thermal continuous transition corresponding to the melting of the three-sublattice order. It is in the 3-state Potts universality class, and the critical temperature vanishes in both limits $h \to 0$ and $h \to 6J$. When $\Delta \neq 0$ and h is small, the state with one spin opposite to the field is selected (the "Y" configuration), and it evolves continuously into the 1/3-plateau up-up-down state at a critical field. Increasing further the field, there is another critical field at which the system leaves the plateau phase to enter a "V" phase in which the down spins progressively align with the field. This is another spin-supersolid phase. Both spin-supersolid phases have been predicted to be separated from the plateau phase by a Berezinskii-Kosterlitz-Thouless (BKT) transition [9]. The development of three-sublattice long-range order coming from high temperature is expected to remain 3-state Potts, except at high field where a single transition into the "V" phase has been predicted.

The very small Δ limit of this model is realized in the cobalt compound $K_2Co(SeO_3)_2$ (KCSO in short), and two teams have recently come up with in-depth experimental investigations of that system that are largely complementary, and that essentially agree when they overlap [12, 13]. The magnetization has a very wide 1/3-plateau and very steep increases from zero magnetization and up to saturation, in qualitative agreement with a very small Δ [12, 13], less than 0.1. High energy inelastic neutron scattering in the plateau phase at 7 T, where the excitations are very clean spin waves, lead to rather precise estimates of the coupling constants $J_z \equiv J = 2.98$ meV and $J_\perp \equiv \Delta J = 0.21$ meV, hence to a ratio $\Delta = 0.07$ [13]. As expected, the overall phase diagram is dominated by a 1/3-plateau phase (see Fig.

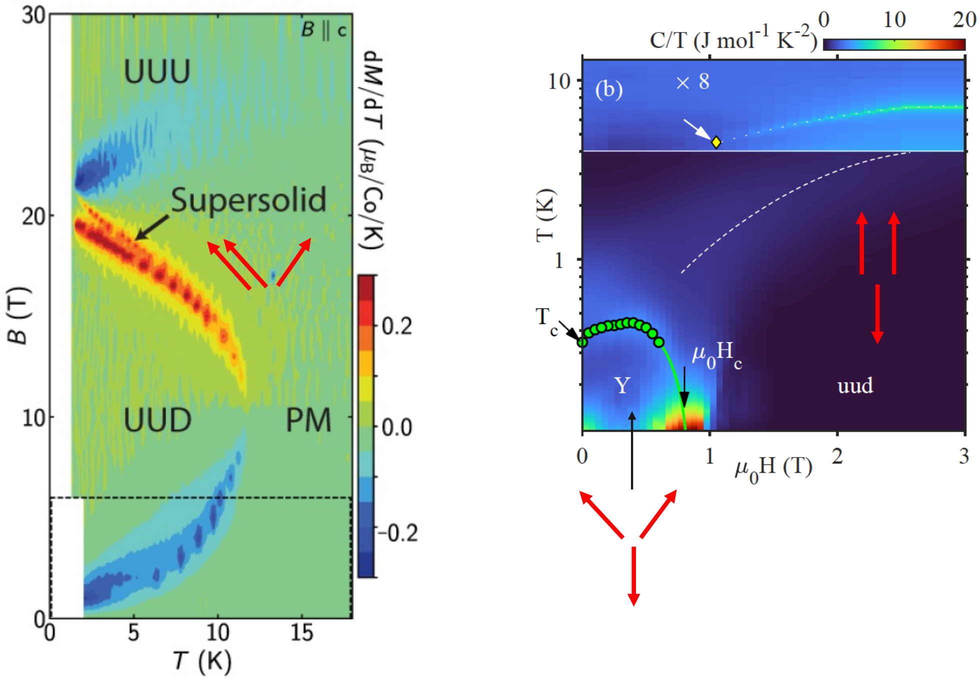

Figure 1: (left) Global phase diagram as deduced from the derivative of the magnetization with respect to the field [13]. The overall shape is typical of the Ising model with a large 1/3 up-up-down plateau that melts at finite temperature in a 3-state Potts transition. The double peak structure of the derivative points to a narrow phase around 20 T that can be interpreted as a spin-supersolid phase where spins adopt the "V" order shown with red arrows. (right) Low-field phase diagram [12]. There is clear evidence of another phase between 0 and 0.8 T. This phase has been suggested theoretically to be another supersolid phase, in which the spins would adopt the "Y" order depicted below the figure, but the presence of long-range order has been challenged by inelastic neutron scattering [12, 13].

1, left panel), as for the Ising model, and the transition is consistent with 3-state Potts.

Let us now concentrate on the low and high field regions of this phase diagram, where transverse exchange is expected to alter the physics qualitatively. At high field, the best evidence of an intermediate phase comes from the double peak structure of the derivative of the magnetization w.r.t. to the field between 18 T and 20 T [13]. In the absence of further evidence, this has been attributed to the presence of a spin supersolid with "V" order.

At low field, the situation is more complex. The peak in the specific heat that marks the 3-state Potts transition looses intensity upon reducing the field and becomes invisible around 1 T [12, 13] (see Fig. 1, right panel). This has been interpreted as an end point [12], but given the absence of a first order transition line continuing that line or crossing it, it seems more likely that the entropy release becomes too small to lead to an observable peak in the specific heat. At much lower temperature, another peak has been detected [12]. Its rounded shape suggests that it could be the trace of the BKT transition into the "Y" shaped supersolid predicted by Gao et al [9]. This is supported by the observation that this line seems to terminate at a quantum critical point at 0.8 T (see Fig. 1, right panel), as expected for the zero-temperature transition between the "Y" phase and the 1/3 plateau.

So far, all the evidence points to the presence of long-range order of "Y" type at low field. However, inelastic neutron scattering results at low field challenge this picture. The first surprising result, although not a definitive problem in itself, is the observation that in the low field part of the 1/3 plateau, at 1.5 T, the excitations are still clear spin waves, but with a very strongly renormalized coupling constant J_z, by about a factor 4 [12], if fitted with linear spin-wave theory. Significant renormalization of the dispersion is known to occur in non-collinear antiferromagnets [14, 15], but not on that scale, and this point definitely requires further investigation.

Even more surprisingly, the excitation spectrum is very anomalous in the putative "Y" phase. First of all, the dispersion, defined by the maximum of intensity at low energy, does not agree with that of linear spin wave theory for the "Y" state. In particular, there is a very well pronounced roton-like minimum at the M point in the Brillouin zone [12] that is absent in linear spin-wave theory (see Fig. 2). Again this might be due to some renormalization since the "Y" state is not collinear, but the magnitude of the effect calls for a careful investigation. Secondly, and maybe more importantly, the dynamical structure factor is not consistent with resolution limited spin waves, but rather looks like a continuum typical of fractional excitations [12, 13]. This is especially clear at the M point, where a continuum is clearly visible in the false color plot of Fig. 2, but even around the K point the excitation branches are much broader that the spin waves at the beginning of the 1/3 plateau at 1.5 T. Could it be that the system is not ordered after all?

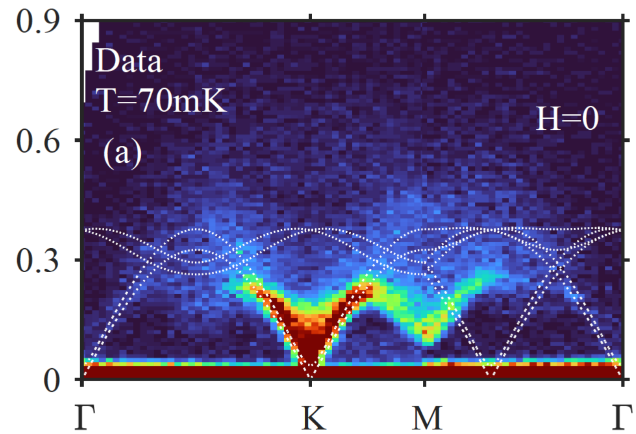

Figure 2: Inelastic neutron scattering in zero field [12] showing that the spectrum is gapless but inconsistent with the spin-wave predictions: There is a deep roton-like minimum at M, and the spectrum consists of a continuum and not of resolution limited spin waves. The linear spin-wave predictions are shown as white dotted lines.

In view of this experimental evidence against long-range order, let us have a critical look at the theoretical evidence in favour of long-range order. The most reliable piece of information comes from the mapping between $\Delta > 0$ and $\Delta < 0$ at small $|\Delta|$ [11]. However, as emphasized by the authors, this proves that there is diagonal order in the bosonic language, i.e. three-sublattice order in the z component of the spins, but *not* that there is off-diagonal long-range order, i.e. long-range order of the components of the spins perpendicular to z, because these correlations are affected by the canonical transformation when going from $\Delta < 0$ to $\Delta > 0$, and they require the introduction of a string in bosonic language. So strictly speaking the mapping between $\Delta > 0$ and $\Delta < 0$ at small $|\Delta|$ implies that there is *partial* order, but not full long-range order.

If the components of the spins perpendicular to z are not fully ordered, the alternative is between a gapped spectrum and exponential correlations, as expected for RVB, or algebraic order with power-law correlations. The experiments on KCSO clearly exclude RVB because there is no gap in the excitation spectrum at the K point. They are however consistent with the Dirac spectrum of an algebraic quantum spin liquid, for which excitations are expected to be gapless but to form a continuum similar to the DesCloizeaux-Pearson continuum of the spin-1/2 Heisenberg chain. Interestingly enough, this would also be consistent with the observation by Ulaga et al [10] that, for small Δ, the anisotropic Heisenberg model on the triangular lattice behaves as the spin-1/2 kagome antiferromagnet.

So, even if RVB is not realized, maybe, as often in his career, Anderson was right about the essential: The Ising-Heisenberg model on the triangular lattice might be a spin liquid after all!

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