Effects of a high magnetic field on the quasi-two-dimensional ferro-antiferromagnet $SrZnVO(PO_4)_2$

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Abstract

Quantum fluctuations in frustrated magnets lead to a possible emergence of new exotic quantum phases. Neutron diffraction was used to observe a first order phase transition in the quasi-two-dimensional ferro-antiferromagnet $SrZnVO(PO_4)_2$ in a magnetic field at 13.65 T. The dispersion relation was determined by using inelastic neutron scattering suggesting that linear spin wave theory is applicable. Albeit the quasi-two-dimensional ferro-antiferromagnetic coupling nature, no spin nematic phase was observed leaving the search for such a material unanswered.

Contents

1 Introduction

The interaction between the magnetic dipoles of a spin system can favour either ferromagnetic ($J < 0$) or antiferromagnetic ($J > 0$) coupling and predicts the magnetic ordering in a solid. Simplified by the symmetry of a given unit cell, a coupling constant J_i is introduced for each pair of individual atoms dependant on the orbital overlap of the wave functions. Dependant on the coupling constants and the underlying lattice an interesting phenomena known as frustration can occur. A frustrated system is characterized by its inability to find a state minimising all the energy contributions arising from the magnetic interactions. In a classical system a strong ferromagnetic coupling constant will favour a fully polarized ferromagnetic state, whereas an antiferromagnetic coupling constant will favour an antiferromagnetic state. This is not the case for quantum magnets, where quantum fluctuations are of importance [1]. For particular quantum systems the interaction is described accurately by the Heisenberg model

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + H \sum S_i^z, \tag{1}$$

where $\langle i, j \rangle$ may denote for example nearest neighbour pairs and $\langle\langle i, j \rangle\rangle$ correspondingly denotes next nearest neighbour pairs. The last term known as the Zeeman term accounts for the effect of an externally applied magnetic field. Interestingly, the classical antiferromagnetic state denoted as $|\mathrm{AF};\, \mathrm{CL}\rangle$ is not an eigenstate of the Heisenberg Hamiltonian [2] but the following identity holds $\mathcal{H}\,|\mathrm{AF};\mathrm{CL}\rangle = \mathcal{E}_{\mathrm{CL}}\,|\mathrm{AF};\mathrm{CL}\rangle + |\alpha\rangle$. In contrast, the ferromagnetic state is an eigenstate of the Hamiltonian which leads to the fact that the actual state occurring at the classical phase boundary has lower energy than the ferromagnetic state. Therefore, the phase boundary is shifted into the ferromagnetic phase when quantum fluctuations are considered. Figure 1 shows several possibilities how the shift may occur in theory.

Based on this argument, a region close to the phase boundary can be expected which is governed by quantum fluctuations and exhibiting an exotic quantum phase (see Figure 1 b) or c)). Realizations of a frustrated system are layered vanadium oxides which are of the structure AA'VO$(PO_4)_2$ (A, A' = Pb, Zn, Sr, Ba). They form quasi-two-dimensional layers of VO_4 pyramids, which are connected through PO_4 tetrahedra in the *ab*-plane [3] as shown in Figure 2 b). It might be tempting to identify this structure as a two dimensional Heisenberg model on a square lattice which, therefore, would exhibit a spin nematic phase at the phase boundary as predicted by theory [4]. However, this is not the case [3] as it is rather more complex as shown in Figure 2 a), where the crystal SrZnVO$(PO_4)_2$ is illustrated.

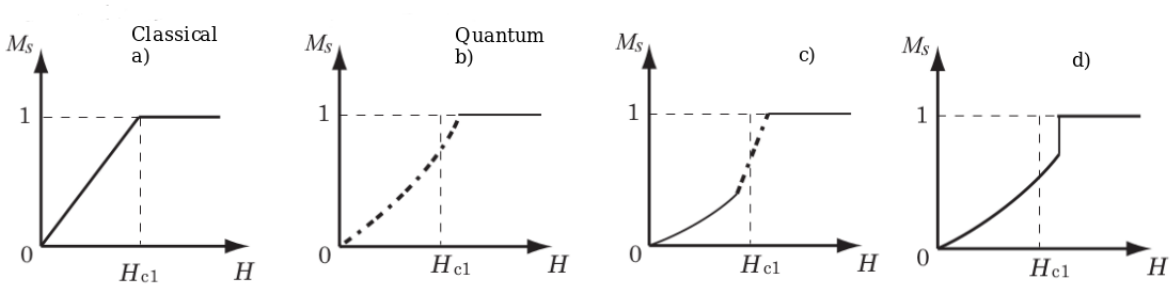

Figure 1: Possible shifts in respect to the classical saturation field H_{c1} of the phase boundary occurring due to the fact that the antiferromagnetic state is not an eigenstate of the Heisenberg Hamiltonian. The solid line corresponds to an ordered state whereas the dashed line represents a possible nontrivial quantum phase. Adapted from [2].

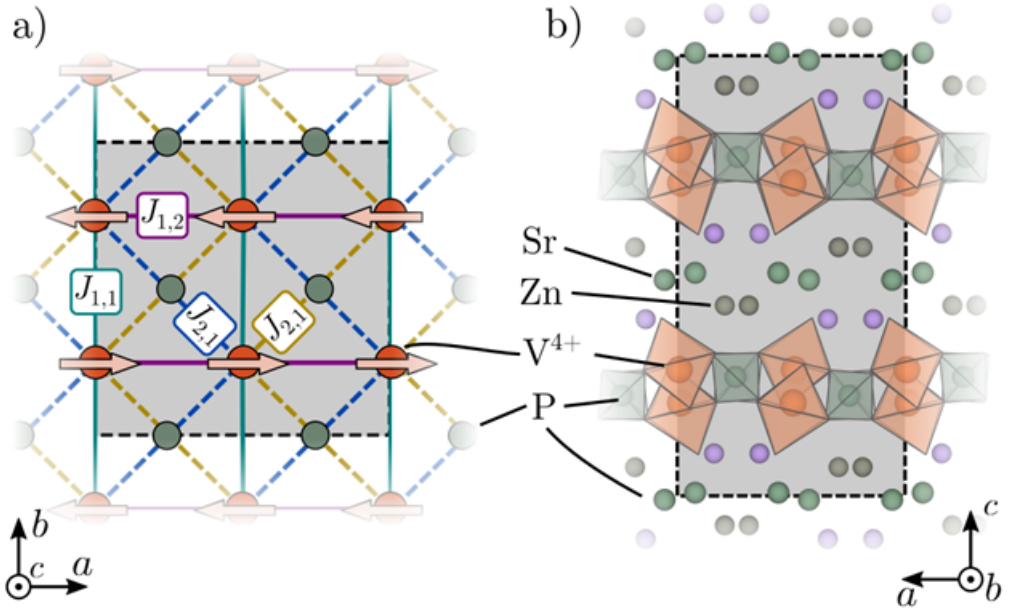

Figure 2: a) In contrast to previous believe, vanadophosphates exhibit a more complex magnetic coupling than a ferromagnetic n.n. and antiferromagnetic n.n.n. coupling on a square lattice. As an example, the magnetic coupling in $SrZnVO(PO_4)_2$ is shown. b) Vanadophosphate layer occurring in crystals of the type $AA'VO(PO_4)_2$, where intermediate atoms are left out for clarity.*

*This figure was created by Florian Landolt, whose consent to publish it in this thesis was granted.

2 Experimental

Periodic modulations in a solid can be detected by scattering of neutrons with a de-Broglie wavelength comparable to the periodicity of the structure of interest. As an example, a crystal with a unit cell length of 10 Å is optimally resolved by neutrons of the energy 0.8 meV which fall in the category of cold neutrons [5]. Fortunately, meV is also the energy scale of elementary excitations in condensed matter systems. Owing the fact that neutrons have a spin and magnetic moment, they experience magnetic dipole interactions with the ions and the electrons, and thus give information about the magnetic ordering in the crystal. In contrast to electrons, neutrons are neutral in charge and can, therefore, penetrate deeper into the crystal and attain bulk information. Overall, neutrons are ideal for measurements of magnetic ordering.

Noting that the interactions between the two-dimensional layers are small[1], the measurements can be simplified dramatically as only excitations within the layers are of relevance. Such a layer can then be aligned with the scattering plane of a three-axis neutron spectrometer (TAS) which in theory provides possibility to measure arbitrary scattering vectors. Limitations however must be taken into account as a change in the scattering vector requires an actual change in the configuration of the instruments, which cannot move to arbitrary angles. The instrument IN12 is an example for a cold neutron TAS and is situated at the Institut Laue-Langevin in Grenoble, where the instrument layout is shown in Figure 3. A change in scattering vector can then be achieved by varying the angles A_1, A_2 up to A_6. Noting that the layers are two dimensional and hence are uniquely described by two degrees of freedom, it is convenient to fix 4 of the 6 angles A. A_1 and A_6 are usually coupled to A_2 and A_5 respectively via the Bragg condition of the monochromator leaving 4 degrees of freedom. A_2 and A_5 determine the length of the initial wave vector $\vec{k_i}$ and final wave vector $\vec{k_f}$ i.e., the energy of the initial neutrons and respectively the energy of the scattered neutrons. In the case of elastic scattering A_2

[1]This argument follows from the consideration of the very similar material $Pb_2VO(PO_4)_2$ [7] and was concluded from a discussion with the author of the referred publication.

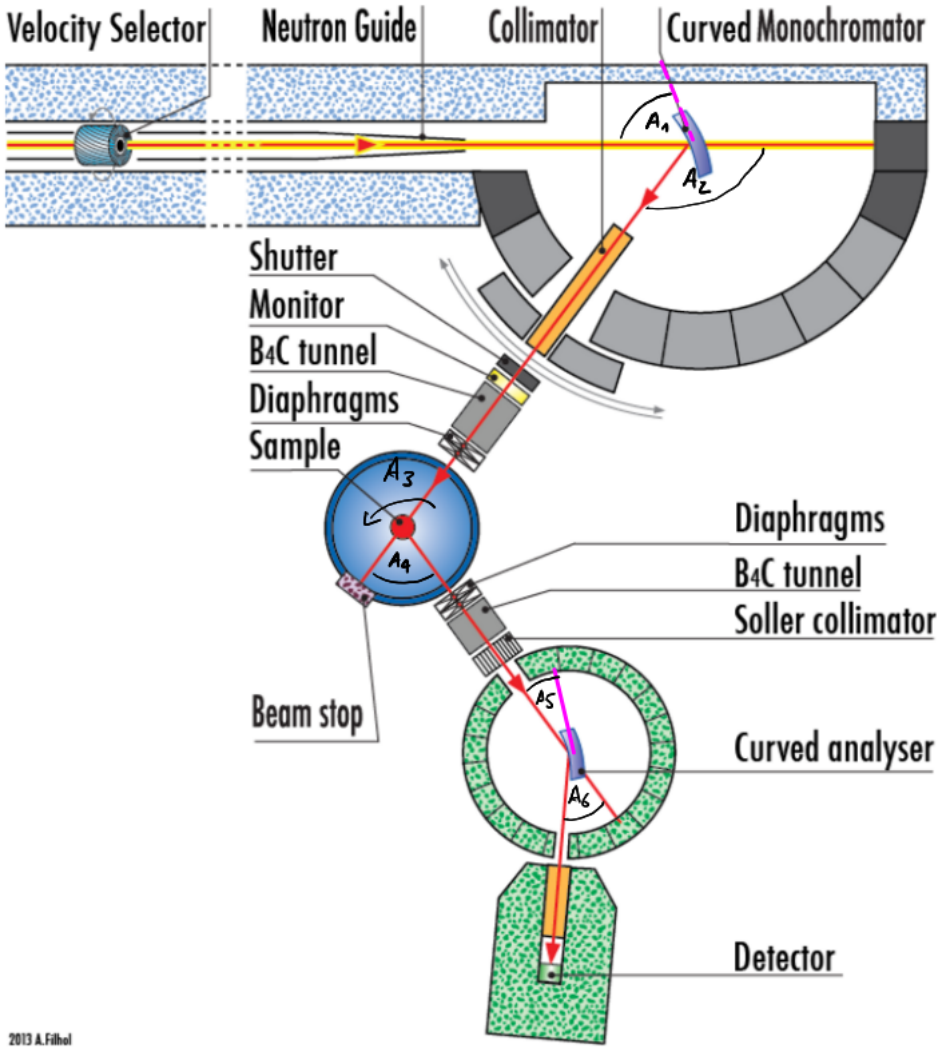

Figure 3: Schematic of the cold neutron three axis spectrometer IN12 at the Institut Laue Langevin in Grenoble. Adapted from [6].

and A_5 remain unchanged and are chosen such that the requirement $\Delta E = |\vec{k_f}| - |\vec{k_i}| = 0$ is fulfilled. This is different when inelastic scattering ($\Delta E \neq 0$) is of interest. Depending on the experimental setup either A_2 or A_5 is fixed and the variation is done in A_5 or A_2 respectively. The scattering vector $\vec{q}$ defined by $\vec{q} = \vec{k_f} - \vec{k_i}$ lies within the scattering plane. Its length is determined by A_4 (θ-scan), whereas the orientation within the plane can be changed via A_3 which is a simple rotation of the sample (ω-scan). Provided that the angles A are within the limits set by the instrument layout, arbitrary $\vec{q}$ and ΔE values can be accessed.

Once the values are set, the monitor counts the number of neutrons per unit time directed onto the sample. This quantity can then be compared to the numbers of neutrons arriving at the detector per unit time. A higher detection rate means that the sample is more likely to form a periodic modulation with the given $\vec{q}$ value of the current configuration than for other $\vec{q}$ values. As an example a Bragg reflection can be studied using an ω-scan, where the acquired data will reveal measurement points which are most likely distributed by a Gaussian curve. As a further example spin waves can be detected by inelastic scattering. Limitations arise at low energies where the elastic line, which has a greater intensity, is situated. Even though possible, measuring the gap of a spin wave is often restricted by the resolution of the configuration of the instruments. On the other hand, static spin density waves can be measured by elastic scattering with increased statistics as the intensity of the Bragg reflections are higher.

Combining a magnetic field to the setup enables the possibility to study the response of the material in a magnetic field while doing diffraction experiments. At IN12 at the Institut Laue-Langevin in Grenoble, a homogeneous magnetic field of 14.9 Tesla and a temperature of 50 mK can be reached simultaneously allowing the measurement presented below.

3 Results

The magnetic Bragg reflection (0,1,0) of the crystal $SrZnVO(PO_4)_2$ was studied using elastic neutron scattering ($k_f = 1.4\text{Å}^{-1}$) in a homogeneous magnetic field, where the resulting data is shown in Figure 4. Throughout the measurement the sample was cooled to a temperature of 50 mK using a dilution refrigerator. For each value of magnetic field an ω-scan was performed and the integrated intensity calculated. Even though the sample was attached to the mount using aluminium wires, the alignment was checked several times throughout the measurement in order to assure that no movement arose due to the magnetic easy axis of the sample which was laying perpendicular to the applied magnetic field. The crystal appears to undergo a first order magnetic phase transition at 13.65 T. Motivated by this result different magnetic Bragg reflections at (1,1,0) and (3,0,0) were expected to appear upon vanishing of the (0,1,0) Bragg reflection. They were, however, not observed for magnetic field values varying between $13,2$ T and 14.9 T.

Based on inelastic neutron scattering, the dispersion relation of a spin excitation in $SrZnVO(PO_4)_2$ was measured at a constant magnetic field of 14.9 T and a temperature of 50 mK. The acquired data is shown in Figure 5. At low energy transfer, the peak arising from elastic scattering covered the inelastic peak explaining why no data of excitations with energy lower than 0.2 meV was retrieved. This limitation accounts for the fact that a possible gap of the spin wave could not be detected by this resolution. The scattering vector (0.5,1.5,0) indicates a crossing of the two branches.

By using vanadium as a reference sample, the resolution of the instrument was determined at zero magnetic field to be 0.1 meV (standard deviation of the Gaussian fit). This value is compared to the width of the dispersion curve shown in Figure 5 and is summarized in the histogram in Figure 6.

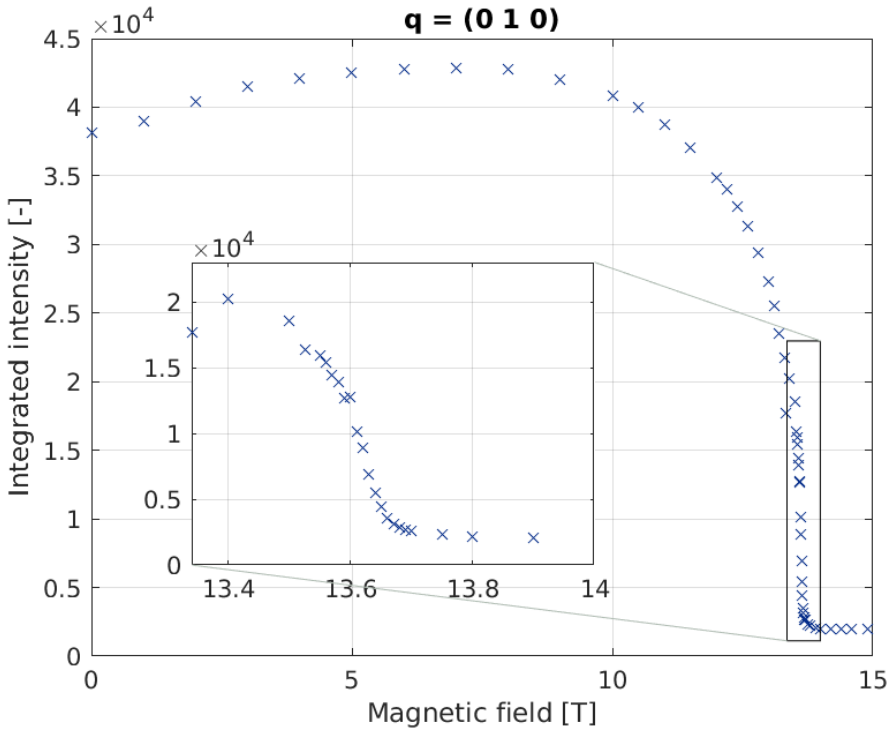

Figure 4: Integrated intensity of the Bragg reflection (0,1,0) as a function of applied magnetic field on the sample. A first order phase transition is observed at around 13.65 T at a constant temperature of 50 mK.

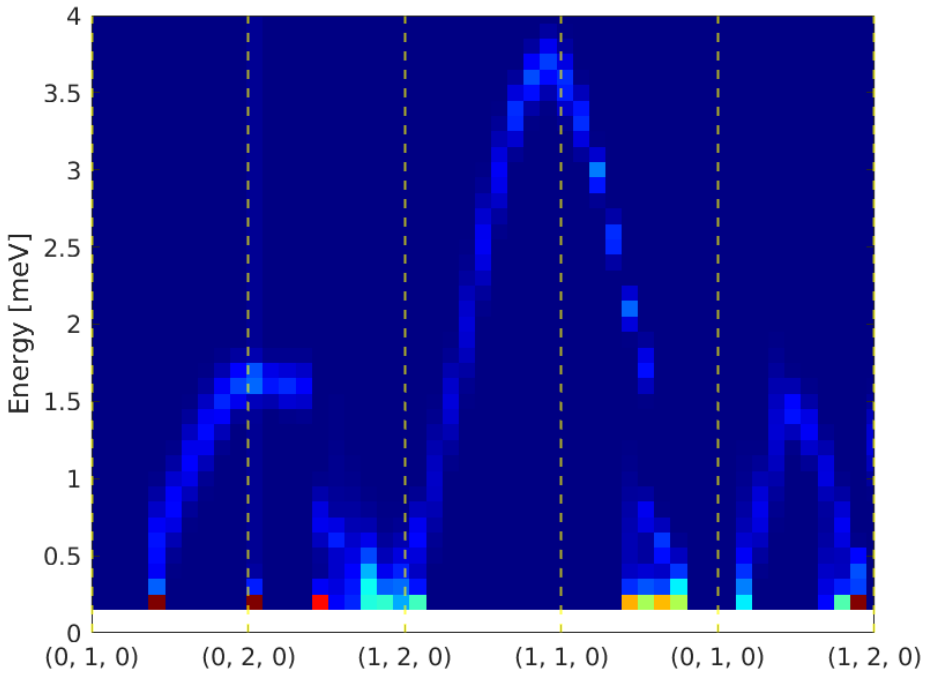

Figure 5: Measured dispersion relation in $SrZnVO(PO_4)_2$ using inelastic neutron scattering at a temperature of 50 mK, where a homogeneous magnetic field of 14.9 T was applied.

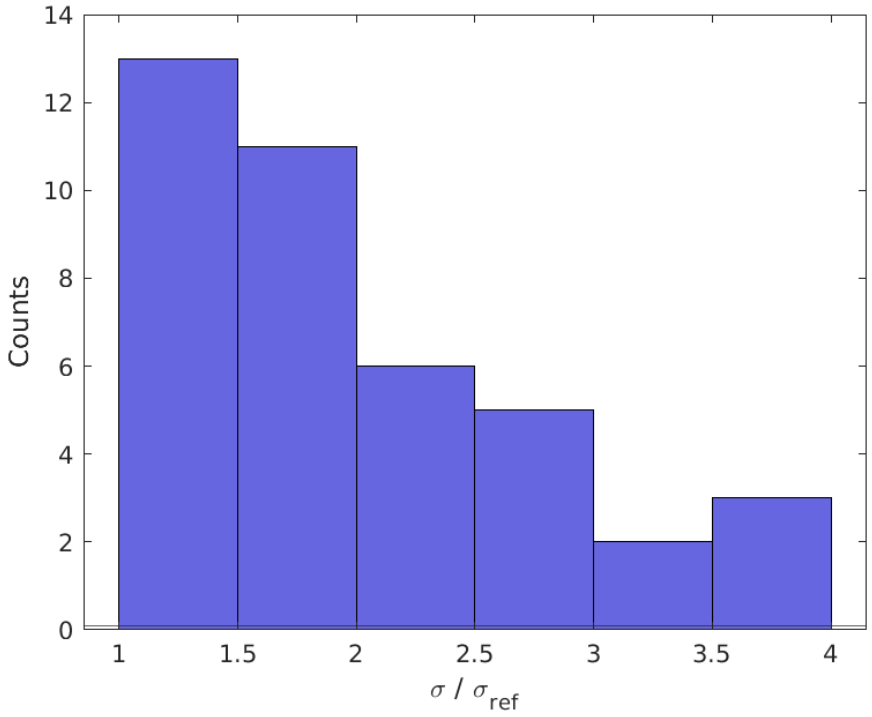

Figure 6: Histogram showing the ratio between the width of the measured dispersion curve and the resolution of the instrument. Measurements with an energy close to the elastic line were not included. The number of bins was found using Sturges' formula [8].

4 Discussion

Within the scope of the acquired data, no sudden rearrangement of magnetic ordering was found replacing the initial (0,1,0) magnetic Bragg reflection at the critical magnetic field of 13.65 T. On the other hand, the transition is of first order indicating that the alignment of the spins along the applied magnetic field is following a more complex mechanism. This is, however, not yet fully understood by the author. In a more sophisticated approach further anisotropy terms in the crystal might be taken into account.

As shown in Figure 6, the width of the dispersion curve is comparable to the resolution of the instrument as it is deviating within an order of magnitude. Therefore, decaying processes of magnons are not significant and linear spin wave theory can be applied. As this was measured in full saturation (14.9 T), the exchange constants J_i can be deduced and will be published in a future article.

5 Conclusion

$SrZnVO(PO_4)_2$ is a two dimensional frustrated quantum magnet described by a Heisenberg Hamiltonian including more than two exchange constants. A neutron scattering measurement revealed that the crystal exhibits a first order phase transition at a magnetic field of 13.65 T and linear spin wave theory can be applied.

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