Quantum fluctuations in high field magnetization of 2D square lattice $J_1$-$J_2$ antiferromagnets.

P. Thalmeier$^1$, M. E. Zhitomirsky$^2$, B. Schmidt$^1$, N. Shannon$^3$

$^1$Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
$^2$Commissariat à l’Energie Atomique, DSM/DRFCM/SPSMS, 38054 Grenoble, France
$^3$H H Wills Physics Laboratory, Bristol BS8 1TL, United Kingdom

(Dated: February 2, 2008)

The $J_1$–$J_2$ square lattice Heisenberg model with spin $S = 1/2$ has three phases with long-range magnetic order and two unconventionally ordered phases depending on the ratio of exchange constants. It describes a number of recently found layered vanadium compounds. A simple means of investigating the ground state is the study of the magnetization curve and high-field susceptibility. We discuss these quantities by using the spin-wave theory and the exact diagonalization in the whole $J_1$–$J_2$ plane. We compare both results and find good overall agreement in the sectors of the phase diagram with magnetic order. Close to the disordered regions the magnetization curve shows strong deviations from the classical linear behaviour caused by large quantum fluctuations and spin-wave approximation breaks down. On the FM side ($J_1 < 0$) where one approaches the quantum gapless nematic ground state this region is surprisingly large. We find that inclusion of second order spin-wave corrections does not lead to fundamental improvement. Quantum corrections to the tilting angle of the ordered moments are also calculated. They may have both signs, contrary to the always negative first order quantum corrections to the magnetization. Finally we investigate the effect of the interlayer coupling and find that the quasi-2D picture remains valid up to $|J_2/J_1| \sim 0.3$.

PACS numbers: 75.10.Jm, 75.30.Ds, 75.60.Ej

I. INTRODUCTION

The search for a quantum spin-liquid — an insulating magnet with a gapless ground state which breaks neither lattice nor spin symmetries — has focused largely on spin-1/2 two-dimensional quantum antiferromagnets (2DQAF’s). In practice, however, most two-dimensional AF spin-1/2 Heisenberg models exhibit either Néel order, or crystals of short-ranged singlet bonds and a finite gap to spin excitations. In a few cases, gapless hidden order states with nematic character arise, but among “realistic” models perhaps only the $S=1/2$ Heisenberg model on a Kagomé lattice remains a serious candidate for a spin-liquid description (see, e.g., references therein).

Perhaps the best studied example of a 2DQAF is the spin-1/2 $J_1$–$J_2$ Heisenberg model, which demonstrates a quantum phase transition from Néel to valance bond solid as a function of the control parameter $J_2/J_1$. A number of layered vanadium compounds have recently been synthesized which are well described by this model. They are of the type $Li_2VOXO_4$ ($X = Si, Ge$) and $AA'VO(PO_4)_2$ ($A, A' = Pb, Zn, Sr, Ba$) consisting of vanadium oxide pyramid layers containing $V^{4+}$ ions with spin $S = 1/2$.

In Refs. [10,11] an extensive analysis of the $J_1$–$J_2$ model, also for finite magnetic field has been given in order to understand physical properties of the above compounds. Both the numerical exact diagonalization (ED) Lanczos method for finite clusters and the analytical spin-wave analysis have been employed. The behaviour of the saturation field as a function of the frustration angle has been studied as a further means of diagnosis of $J_1$–$J_2$ compounds. It was found that close to the disordered regime with $J_1 < 0$ it is determined by an instability of two spin excitations, indicating indeed that in this regime the ground state may be of a spin-nematic type. This leads us to expect that the magnetization itself should also be anomalous in this regime with a large effect from quantum fluctuations. So far quantum corrections to the magnetization curve in the spin-wave theory have only been considered for the nonfrustrated square-lattice antiferromagnet ($J_2=0$).

In this work we give a systematic investigation of the magnetization and the high-field susceptibility for the general 2D square lattice $J_1$–$J_2$ model. Our goal is to investigate how the quantum corrections on these quantities depend on the degree of frustration, especially close to the disordered phases. The effect of interplane coupling will be also considered.

The low scale of exchange interactions which are of order 10 K in the $Li_2VOXO_4$ and $AA'VO(PO_4)_2$ vanadates mean that magnetization measurements are relatively easy to perform. It is therefore hoped that the analysis with the theory developed here gives an additional criterion to determine the frustration ratio $J_2/J_1$ in a specific compound. In Sec. II we briefly introduce the model. Its high field properties like magnetization and susceptibility are investigated in Sec. III. They are obtained from ED Lanczos calculations and first as well as second order spin-wave theory and a comparison is given. We also evaluate the contributions of interlayer coupling in Sec. IV and in Sec. V we finally give a discussion and conclusion.
II. THE J1-J2 MODEL AND ITS PHASES

The 2D square lattice spin-1/2 Heisenberg model in an external magnetic field $H$ is given by

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle} S_i S_j + J_2 \sum_{\langle ij \rangle_2} S_i S_j - h \sum_i S_i^z, \quad (1)$$

where $J_1$ and $J_2$ are the two exchange constants between the first and the second neighbors on a square lattice, respectively. As in Refs. 10,11 the exchange parameters are defined per exchange bond. Furthermore we use the convention $\hbar = g\mu_B H$ ($g$ = gyromagnetic ratio, $\mu_B$ = Bohr magneton). The phases in zero field are best characterized by introducing equivalent parameters

$$J_c = (J_1^2 + J_2^2)^{\frac{1}{4}}, \quad \phi = \tan^{-1}(J_2/J_1), \quad (2)$$

or $j = \tan \phi = J_2/J_1$. The angle $\phi$ determines the extent of magnetic frustration in the model.

Three classical magnetic ground states are possible depending on $\phi$: namely ferromagnet (FM), Néel antiferromagnet and collinear antiferromagnet (CAF). They have been extensively discussed in Ref. 11. The effect of exchange frustration leading to enhanced quantum fluctuations is strongest at the classical phase boundaries where the CAF phase joins the NAF or FM phase (see insets of Fig. 1). In fact in these regions they are believed to destroy long-range magnetic order and establish two new partially ordered states, namely a columnar dimer state with a spin gap at CAF/NAF boundary and a gapless spin nematic state at the CAF/FM boundary. It can be seen already within spin wave approximation that the magnetic order breaks down in this regime since the sublattice moment reduction due to quantum fluctuations diverges close to the two boundary regions.

III. HIGH FIELD PROPERTIES OF THE J1-J2 MODEL

The determination of the frustration ratio $J_2/J_1$ or angle $\phi = \tan^{-1}(J_2/J_1)$ is of foremost importance to characterize a given square lattice magnetic compound such as the SiO$_4$ and PO$_4$ vanadates mentioned in the introduction. The available experimental methods to reach this objective have been extensively discussed in Ref. 10. However, the thermodynamic zero-field methods like evaluating the heat capacity and the magnetic susceptibility are too ambiguous to locate $\phi$ in the NAF or CAF sector of the phase diagram. Additional information may be obtained from investigating high-field properties, for example from saturation fields as, e.g., determined from the magnetocaloric effect.

The analysis of the high field magnetization itself as function of frustration angle $\phi$ is also promising. From the simple n.n. Heisenberg AF ($J_2 = 0$ or $\phi = 0$) it is known from analytical work that deviations from the classical linear magnetization curve due to quantum fluctuations are to be expected. This is also concluded from numerical calculations. A systematic study of magnetization curves for the $J_1$-$J_2$ model is however lacking. Since the method is of experimental importance due to its relative simplicity we consider it worthwhile to investigate this problem in detail for the antiferromagnetic phases of the $J_1$-$J_2$ model. For this purpose we will use both analytical spin wave methods similar to Ref. 18 and numerical Lanczos methods for exact diagonalization of finite clusters. Our notation will be close to the one used in Ref. 11.

A. Magnetization from numerical $(T = 0)$ Lanczos results for $J_1$-$J_2$ clusters

We have numerically diagonalized the Hamiltonian of the $J_1$-$J_2$ model for cluster sizes of 16, 20, and 24 sites applying the finite-temperature Lanczos method as described in Ref. 11 and references cited therein. In the limit $T \to 0$, our results are identical to the standard zero-temperature implementation of the algorithm, i.e., the evaluation of the partition function reduces to the determination of ground-state expectation values. The clusters of size 16 and 20 are (regular and tilted) squares, the 24-site cluster is a rectangle. All three clusters tile the infinite lattice such that, with periodic boundary conditions, compatibility with the three classically ordered ground states is preserved.

The zero-temperature field dependence of the magnetization $m = \langle S^z \rangle$ has been calculated for the whole phase diagram except the FM region, where the saturation field $h_s$ vanishes. Following Ref. 20, Fig. 1 shows the normalized magnetization $m/m_{\text{sat}}$ ($m_{\text{sat}} \equiv S$) for selected values of the frustration angle $\phi$ in the classically ordered antiferromagnetic phases and in the two disordered regimes of the phase diagram. The magnetic field is normalized to the saturation field determined by exact diagonalization. This is identical to the classical saturation field for positive (antiferromagnetic) $J_1$. For negative (ferromagnetic) $J_1$, a $\Delta S = 2$ two-magnon instability determines the saturation field for the finite-size systems considered here, which occurs at slightly higher field values than the one-magnon instability.

The field dependence of the magnetization at $T = 0$ for a finite-size system is a sequence of finite steps. The solid line in the left panel of Fig. 1 shows the magnetization curve of the 24-site cluster for $\phi = -0.21 \pi$, which is in the Néel phase, the dashed line shows the same for $\phi = 0.75 \pi$ in the collinear phase. In the right panel, the solid line shows the field dependence for $\phi/\pi = 0.17$ (columnar dimer phase), the dashed line for $\phi/\pi = 0.84$ (spin-nematic phase). The symbols in the plots denote the midpoints of the horizontal and vertical line segments of the magnetization steps; circles label the 24-site data, diamonds the 20-site data, and squares the 16-site data. (For the smaller cluster sizes, the step functions are not
The angles $\phi/\pi = -0.21$ and 0.75 for the NAF and CAF in the left plot are chosen such that they correspond to the experimental findings for the compound SrZnVO$_2$(PO$_4$)$_2$. For these values, the midpoints of the magnetization steps form a smooth function. This can generally be observed for any frustration angle located inside the magnetically ordered regimes, be it NAF or CAF. In contrast, for $\phi/\pi = 0.17$ (solid symbols and line in the right plot) and $\phi/\pi = 0.85$ (open symbols and dashed line), the data points scatter much more and do not give rise to a smooth field dependence. Furthermore, in the columnar dimer phase ($\phi/\pi = 0.17$) the well-established half-magnetization plateau that appears at $m = \frac{1}{2} m_{\text{sat}}$ is observed. The data in the right panel of Fig. 1 indicate possibility of yet another plateau at $m = \frac{1}{2} m_{\text{sat}}$, though a careful examination of finite-size effects is necessary to make a final conclusion.

In the whole phase diagram, quantum effects lead to negative corrections: the exact magnetization curve $m(h)$ lies always below the corresponding classical value, which is a consequence of the lowering of the ground state energy of a quantum antiferromagnet compared to its classical counterpart $m_{\text{sat}}$.

### B. Spin wave excitations in an external magnetic field

A standard Holstein-Primakoff approximation of Eq. (1) and a subsequent Bogoliubov transformation leads to the harmonic spin wave Hamiltonian:

$$H = N E_0 + N E_{zp} + \sum_{\lambda k} \epsilon_{\lambda k}(h) a_{\lambda k}^\dagger a_{\lambda k}, \quad (3)$$

where $a_{\lambda k}$ are magnon operators that obey bosonic commutation rules. The $\epsilon_{\lambda k}(h)$ denote the spin wave dispersion of branch $\lambda = \pm$ as defined in the appropriate NAF or CAF magnetic Brillouin zone (BZ). It is given by

$$\epsilon_{\pm k}(h) = \sqrt{A_k(h) \pm C_k(h)^2 - B_k(h)^2}. \quad (4)$$

Here $A_k(h)$ is the intra- and $B_k(h)$, $C_k(h)$ are the inter-sublattice couplings given below. The ground state energy is composed of a classical part ($E_0$) obtained from the mean field approximation to Eq. (1) and a part due to zero point fluctuations of spins ($E_{zp}$). The former is given by

$$E_0 = - h \langle S_\parallel \rangle + a_\parallel \langle S_\parallel \rangle^2 - a_\perp \langle S_\perp \rangle^2, \quad (5)$$
FIG. 2: Magnetization curves $\mu/\mu_B = gm = m/S$ for various frustration angles in the antiferromagnetic or disordered sectors with an offset of 0.2 applied. Symbols are obtained from Lanczos magnetization data for $N = 16$ (squares), 20 (diamonds), 24 (circles, dots) size clusters using the Bonner-Fisher construction\textsuperscript{20}. Lines are obtained from first (full) or second (dotted) order spin wave calculations of Sects. III C and III F. Angles $\phi/\pi = 0.75, -0.21$ correspond to the possible CAF or NAF values of the Sr compound. Magnetization curves strongly differ in the extent of nonlinear deviation from the classical curve which corresponds to $\phi/\pi = -0.5$. Furthermore $\phi/\pi = 0.41, -0.17$ are values which are deeply within the CAF or NAF regions and overall agreement of spin wave and Lanczos calculations is good. It is less so on the CAF side where the magnetization changes in steps of $\Delta S_z = 2$ leading to a larger finite size scattering. The values $\phi/\pi = 0.75, 0.17$ correspond to regions close to or within the nonmagnetic sectors. Close to the CAF/FM boundary the first order spin wave results overemphasize the nonlinear behaviour and become unstable at very low fields. Close to the CAF/NAF boundary the numerical data exhibit a plateau at $m/S = \mu/\mu_B = 0.5$ which would require a separate analysis. The plateau was first reported in Ref. \textsuperscript{22}. The inset shows the position of plotted $\phi$ values in the phase diagram. Second order spin wave results are discussed in Sect. III F.
where $\langle S_i \rangle = S \cos \frac{\theta_c}{2}$, $\langle S_\perp \rangle = S \sin \frac{\theta_c}{2}$ and $\theta_c/2$ is the classical field induced canting angle of AF moments counted from the $z$-axis which is chosen parallel to the applied field $h$ (here $\parallel \equiv z$ and $\perp \equiv x, y$). The coefficients in Eq. (5) are given by

$$a_\parallel = \frac{z}{2}(J_1 + J_2) , \quad a_\perp = \frac{z}{2}(J_1 - J_2) , \quad (\text{NAF})$$

$$a_\perp = \frac{z}{2}J_2 , \quad (\text{CAF})$$

where $z = 4$ is the coordination number. The classical canting angle $\theta_c/2$ of moments is obtained minimizing $E_0$ which leads to

$$\cos \frac{\theta_c}{2} = \frac{h}{h_s} , \quad h_s = 2S(a_\parallel + a_\perp) , \quad (7)$$

where $h_s$ is the classical saturation field. For $h \geq h_s$ the moments are fully polarized, i.e., all are ferromagnetically aligned parallel to $h$. Explicitly we have $h_s = 2zS J_1$ (NAF) and $h_s = zS(J_1 + 2J_2)$ (CAF).

The zero point energy due to quantum fluctuations is obtained as

$$E_{zp} = \frac{1}{2N} \sum_{\lambda k} \left[ \epsilon_\lambda k(h) - A_k \right] . \quad (8)$$

For the CAF and NAF phases it is always negative and vanishes in the FM or fully polarized ($h = h_s$) phase where the ground state and spin wave states are exact eigenstates with a dispersion $\epsilon_\lambda k \equiv A_k$. The sublattice couplings which determine the spin wave dispersion in the canted state were derived in Ref. [11] and are given here for completeness. Defining

$$A_k = S a_k , \quad B_k(h) = S b_k \sin^2 \frac{\theta_c}{2} , \quad C_k(h) = S c_k \cos^2 \frac{\theta_c}{2} , \quad (9)$$

we have in the NAF and CAF phases corresponding to wave vectors $Q = (\pi, \pi)$ and $Q = (\pi, 0)$, respectively:

$$a_k = 4[J_1 - J_2(1 - \tilde{\gamma}_k)] , \quad (\text{NAF})$$

$$c_k = -b_k = 4J_1 \gamma_k , \quad (10)$$

$$a_k = 2[2J_1 + J_1 \gamma_y] , \quad (\text{CAF})$$

$$c_k = -b_k = 2[J_1 + 2J_2 \gamma_y] \gamma_x ,$$

where the geometric structure factors are defined by

$$\gamma_k = \frac{1}{2}(\cos k_x + \cos k_y) ,$$

$$\tilde{\gamma}_k = \cos k_x \cos k_y , \quad \gamma_x = \cos k_x , \quad \gamma_y = \cos k_y . \quad (11)$$

Due to the symmetry properties $A_{k+Q} = A_k$, $B_{k+Q} = -B_k$ and $C_{k+Q} = -C_k$ we have the identity $\epsilon_{\lambda,k}(k+Q) = \epsilon_{\lambda,k}(k)$. Then, instead of summing over two spin wave branches in the magnetic BZ of NAF or CAF, we may restrict to one mode only, e.g. $c_k \equiv \epsilon_{+,k}$ and sum over the whole paramagnetic BZ in the expression for $E_{zp}$ and similar ones. Using this convention the spin wave branch index $\lambda$ will be omitted in the following. Using Eqs. (10) and (11) the spin wave energies may be written as

$$c_k(h) = S(a_k + c_k) \frac{1}{2}(a_k + c_k \cos \theta_c) \frac{1}{2} , \quad (12)$$

for both NAF and CAF cases with $a_k$ and $c_k$ given in Eq. (10).

C. Magnetization from first order spin wave quantum corrections

The zero temperature magnetization is given by the field derivative of the total ground state energy:

$$m = m_0 + m_{zp} = -\frac{\partial E_0(h)}{\partial h} - \frac{\partial E_{zp}(h)}{\partial h} , \quad (13)$$

where the first term is the linear classical part and the second one the (negative) correction due to quantum fluctuations included up to first order in $1/S$. We can write

$$\frac{\partial c_k(h)}{\partial h} = \frac{1}{c_k(h)} 2C_k \left[ (A_k + C_k) - B_k \right]$$

$$= \frac{2S}{h_s} c_k \left[ \frac{a_k + c_k}{a_k + c_k \cos \theta_c} \right] \frac{1}{2} \cos \frac{\theta_c}{2} . \quad (14)$$

This finally leads to a total magnetization, including the first order quantum corrections:

$$m = S \frac{h}{h_s} \left[ 1 - \frac{1}{h_s} N \sum_k c_k \left( \frac{a_k + c_k}{a_k + c_k \cos \theta_c} \right) \frac{1}{2} \right] , \quad (15)$$

where on the r.h.s. the classical value of $\theta_c$ given by $\cos(\theta_c/2) = h/h_s$ has to be used. Because $h_s = 2S(a_\parallel + a_\perp)$ the second term in Eq. (15) is formally a $1/S$ correction to the linear classical term $m_0 = S(h/h_s)$. Explicitly, using Eqs. (10,11) we have for the NAF case a magnetization depending on field strength and frustration angle according to
The zero-field susceptibility tends to zero indicating the breakdown of the \(1/S\) approximation. Right panel: Zero-field normalized sequence for \(h = 0\). Note the divergence close to the result first given in Ref. 18.

The positive value (full line, symbols) but they diverge on approaching the CAF/NAF boundary around \(\phi/\pi \simeq 0.15\).

\[
\chi = \frac{J^2}{h^2} \left[ 1 - \frac{1}{2SN} \sum_k \gamma_k \left( \frac{1 + \gamma_k - j(1 - \gamma_k)}{1 + \gamma_k \cos \theta_s - j(1 - \gamma_k)} \right) \right],
\]

where we used \(j = \tan \phi = J_2/J_1\). In this expression the \((1/S)\) character of the quantum correction becomes manifest. For the simple NAF with \(j = 0\) we reproduce the result first given in Ref. 18.

A similar but more complicated expression may be given for the CAF phase. Defining \(\delta_k = (1/2)(\cos kx - \cos ky)\) in addition to Eq. 11 we obtain

\[
m = S \frac{h}{h_s} \left[ 1 - \frac{1}{2SN(j + \frac{1}{2})} \sum_k \frac{1}{2}(\gamma_k + \delta_k) + j\gamma_k \left( \frac{j(1 + \gamma_k) + \gamma_k}{j(1 + \gamma_k \cos \theta_s) + \gamma_k \cos^2 \frac{\theta_s}{2} - \delta_k \sin^2 \frac{\theta_s}{2} \right) \right].
\]

Note that the special CAF case with \(J_1 = 0\) (\(j = \infty\)) is equivalent to the simple NAF case \(J_2 = 0\) (\(j = 0\)) in Eq. 10. This may be seen by applying a \(k\)-coordinate rotation by \(\pi/4\) in Eq. 17.

Naturally, the above expressions should be primarily valid deep inside the NAF and CAF regions where the staggered moments are large. Close to the boundaries quantum fluctuations grow and destroy the magnetic order, then corrections to \(m(h)\) starting from the ordered state and expanded in orders of \(1/S\) are no longer appropriate.

The combined analytical spin wave and numerical Lanczos results for the magnetization are shown in Fig. 2. One may indeed see that the agreement of both is good deep inside the NAF \((\phi/\pi = -0.21, -0.17)\) and CAF \((\phi/\pi = 0.41)\) sectors. On the other hand close to the classical phase boundaries \((\phi/\pi = 0.17, 0.75)\) discrepancies appear. For the former case our and previous\(^{22}\) Lanczos results indicate appearance of the one-half magnetization plateau with \(m = \frac{1}{4}m_{sat}\) close to \(h/h_s = 0.5\). (Such a plateau has been also found in the equivalent classical model at finite temperatures\(^{22}\) For the latter the magnetization curve becomes very nonlinear due to quantum fluctuations and \(m(h)\) becomes negative at low fields (upper dashed line) indicating the breakdown of the \(1/S\) expansion. This means that the CAF state becomes unstable and a new (spin nematic) order parameter will be realized in a finite sector around \(\phi/\pi = 0.85\) \((J_2/J_1 \sim -0.5)\). Generally, one may say that quantum corrections leading to nonlinear magnetization will be considerable larger on the ferromagnetic \((J_1 < 0)\) side of the CAF sector. This will be further discussed in Sec. 5.
D. First order quantum corrections to the magnetic susceptibility

In the spin-wave approximation the magnetization curve for the simple AF exhibits a logarithmic singularity of the slope close to the saturation field $h_s$ as shown in Refs. 18,21. This should be more easily visible in the high field susceptibility. Furthermore for $\phi$ approaching a classical phase boundary the low field susceptibility has to vanish, which signifies the instability of the order parameter. For these reasons we found it useful to study the magnetic susceptibility $\chi(h, \phi) = \partial m(h, \phi) / \partial h$ as function of the frustration angle $\phi$ in addition to the magnetization.

From Eq. (13) we obtain

$$\chi(h, \phi) = \chi_0 + \chi_{zp} = S \frac{1}{h_s} - \frac{1}{2N} \sum_k \left( \frac{\partial^2 c_k(h)}{\partial h^2} \right).$$  \label{eq:18}

From Eq. (12) and using $(\partial A_k / \partial h) = 0$ we arrive at

$$\frac{\partial^2 c_k(h)}{\partial h^2} = \left( \frac{\partial c_k}{\partial h} \right) \left[ \frac{1}{h} - \frac{1}{c_k(h)} \left( \frac{\partial c_k(h)}{\partial h} \right) \right].$$  \label{eq:19}

Inserting this in Eq. (18) and using Eq. (13) leads to a general expression of the normalized susceptibility $\chi_n(h) = h_s \chi(h)$ according to

$$\chi_n(h, \phi) = S \left[ 1 - \frac{1}{S} (\Delta \chi_n^{(a)}(h, \phi) - \Delta \chi_n^{(b)}(h, \phi)) \right],$$  \label{eq:20}

where the terms $\sim 1/S$ in brackets are the quantum corrections of $\chi_{zp}^n$ to the constant classical value $\chi_0^n$. We obtain from Eqs. (18,19):

$$\Delta \chi_n^{(a)}(h, \phi) = \frac{S}{h_s} \frac{1}{N} \sum_k c_k \left( \frac{a_k + c_k}{a_k + c_k \cos \theta_c} \right)^{\frac{1}{2}},$$  \label{eq:21}

$$\Delta \chi_n^{(b)}(h, \phi) = 2 \frac{S}{h_s} \frac{1}{h_s} \frac{1}{N} \sum_k c_k^2 \left( \frac{a_k + c_k \cos \theta_c}{1 + c_k \cos \theta_c} \right)^{\frac{1}{2}}.$$

For the NAF case this may be explicitly evaluated as

$$\Delta \chi_n^{(a)}(h, \phi) = \frac{1}{2N} \sum_k \gamma_k \left( \frac{1 + \gamma_k - j(1 - \tilde{\gamma}_k)}{1 + \gamma_k \cos \theta_c - j(1 - \gamma_k)} \right)^{\frac{1}{2}},$$

$$\Delta \chi_n^{(b)}(h, \phi) = \left( \frac{h}{h_s} \right)^2 \frac{1}{N} \sum_k \gamma_k^2 \left( \frac{1 + \gamma_k - j(1 - \tilde{\gamma}_k)}{1 + \gamma_k \cos \theta_c - j(1 - \gamma_k)} \right)^{\frac{1}{2}}.$$  \label{eq:22}

Obviously only the first part $\Delta \chi_n^{(a)}$ contributes to the $(1/S)$ corrections of the zero-field susceptibility. It may also be directly obtained by differentiation of $m(h)$ in Eq. (10). For the CAF case similar expressions for $\Delta \chi_n^{(a,b)}(h, \phi)$ may be derived by making analogous substitutions in the integrals and their prefactors as done in Eqs. (16) and (17).

The typical field dependence of the susceptibility is shown in Fig. 3. For $\phi/\pi = -0.49$ close to the NAF/FM boundary one obtains nearly the classical constant value $\chi_n = S$ because in the FM sector quantum fluctuations are not present, they are gradually turned on when $J_1$ becomes positive and $\phi$ moves into the NAF sector. This can be clearly seen from the various curves in Fig. 3(left). For an angle $\phi/\pi = 0.1$ frustration becomes large and quantum fluctuations are close to destroying the NAF order. Accordingly the zero-field susceptibility is close to becoming negative where the spin-wave theory breaks down. One also notes the upturn in the susceptibility just below the critical field coming from the logarithmic singularity of the magnetization. The singularity becomes stronger when the strongly frustrated point $j = 1/2$ is approached.

E. Quantum corrections of the moment canting angle

The angle $\theta_c$ between canted AF ordered moments has so far been given in classical approximation from minimizing only $E_0(\theta_c, h)$. Its quantum corrections of first order in $(1/S)$ may be computed by minimizing the total energy $E_0(\theta_c, h) + E_{zp}(\theta_c, h)$, keeping $\theta_c$ also as variable in the zero point energy. For the unfrustrated Néel AF this correction is small, however, as we shall see in the following it may be of considerable size close to the classical boundaries of NAF and CAF phases where frustration effects are large.

The equilibrium condition including quantum corrections is given by

$$\frac{\partial E_0}{\partial \theta_c} + \frac{1}{2N} \sum_k \frac{\partial c_k}{\partial \theta_c} - \frac{A_k}{\partial \theta_c} = 0.$$  \label{eq:23}

Now $\theta_c$ is to be treated as a variable present in $c_k$ and $A_k$. The form of $A_k$ in Eqs. (12,13) has already the classical angle of Eq. (7) substituted. Its general form is $A_k = S \bar{a}_k$ with

$$a_k = -4J_2(1 - \tilde{\gamma}_k) - 4J_1 \cos \theta_c + h \cos \frac{\theta_c}{2},$$  \label{eq:24}
FIG. 4: Comparison of \((1/S)\) quantum corrected canting angle \(\theta'_c\) (left) and magnetization (right) for two values in the NAF sector. It is seen that first order quantum corrections always reduce the magnetization with respect to the classical value according to Eq. (15). For \(\cos(\theta'/2)\) however the corrections may have both signs. They are small and negative for \(\phi\) deep in the NAF sector while they are positive closer to the NAF/CAF boundary.

for the NAF case. Likewise for CAF sector one obtains

\[
a_k = -2J_1(1 - \gamma_y) - 2(J_1 + 2J_2)\cos\theta_c + h\cos\theta_c/2.
\]

(25)

Evaluating the derivatives of \(\epsilon_k\) and \(A_k\) with respect to \(\theta_c\) and solving the equilibrium Eq. (23) we obtain the canting angle \(\theta'_c\) renormalized by quantum fluctuations:

\[
\cos\theta'_c/2 = \cos\theta_c/2 \left\{1 - \frac{1}{h_s} \frac{1}{N} \sum_k \left[ \frac{a_k^{(1)}(a_k + c_k\cos\theta_c/2)}{(a_k + c_k)^2(a_k + c_k\cos\theta_c/2)} - a_k^{(1)} \right] \right\}.
\]

(26)

Here \(\cos\theta'_c/2 = h/h_s\) is the classical canting angle and \(a_k^{(1)} = -(h_s/2S)\) is the coefficient of the second term in Eqs. (24, 25). Again, because of \(h_s = 2S(a_\parallel + a_\perp)\) the term \(\sim 1/h_s\) in Eq. (26) has to be considered as a \((1/S)\) correction to the canting angle. Therefore in the sum on the r.h.s. the classical values for \(\theta_c\) has to be used which leads again to the intra-sublattice coupling \(a_k\) as given by Eq. (10).

The general solution in Eq. (26) is valid for both AF phases. In the NAF case it leads to the explicit result

\[
\cos\theta'_c/2 = \cos\theta_c/2 \left\{1 - \frac{1}{2S} \frac{1}{N} \sum_k \left[ \frac{\gamma_k^2 + \gamma_k\sin^2\theta_c/2 - 1 + j(1 - \gamma_k)(1 - \gamma_k)}{(1 + \gamma_k - j(1 - \gamma_k))(1 + \gamma_k\cos\theta_c - j(1 - \gamma_k))^2} + 1 \right] \right\}.
\]

(27)

For the simple AF (\(j = 0\)) one recovers the expression given in Ref. [18]. A similar explicit expression for the CAF case may be derived but it is too unwieldy to be given here. For the numerical calculation of \(\cos\theta'_c/2\) Eq. (26) may be used as well.

The field dependence of renormalized moment canting given by \(\cos\theta'_c/2\) in comparison with the normalized magnetization \(m(h)/S\) is shown in Fig. 4. Generally the quantum corrections to the canting angle are quite small deep inside the AF sectors. As for the magnetization they become larger when approaching a phase boundary. Interestingly however, they may have different signs for the
former (left panel) while they must always be negative for the latter (right panel). The positive correction to \( \cos \frac{\phi}{2} \) appears in the vicinity of the NAF/CAF boundary.

The quantum corrections to magnetization and canting angle may be used to obtain the correction to the moment size. Classically we have \( S = m_0 / \cos \frac{\phi}{2} \). If we use a similar definition including the quantum corrections we have \( \langle S \rangle = m / \cos \frac{\phi}{2} \) for the renormalized moment. The change of moment size defined by \( \delta S = S - \langle S \rangle \) is then given by \( \delta S = S - m / \cos \frac{\phi}{2} \). Note however that in this relation the quantum correction to \( S \) formally contains effects of arbitrary order in \( 1/S \) even if \( m \) and \( \cos \frac{\phi}{2} \) are only corrected in order \( 1/S \). Close to the nonmagnetic regions when \( m \) approaches zero \( \delta S / S \) becomes unity, i.e., the staggered moment is destroyed by the quantum fluctuations.

Here the first line is equal to the sum of \( E_0 + E_{zp} \) while the second and third line constitute the second order in \( 1/S \) spin-wave contribution. In this term the six constants are given by two-dimensional momentum integrals:

\[
E_{g.s.}/N = -2(J_1 - J_2)S(S + 1) - \frac{\hbar^2}{16J_1} + \frac{1}{2N} \sum_k \varepsilon_k
- 2J_1 [(n - \delta_1)^2 + n_1(n_1 - \delta)] + 2J_2 [(n - n_2)^2 + \delta_2 (\delta_2 - \delta)]
+ 4J_1 \cos^2(\theta_c/2) [(n - \frac{1}{2} \delta)(\delta_1 + n_1) - 2\delta_1 n_1] - J_1 \sin^2 \theta_c \frac{1}{3N^2} \sum_{k,q} F(k, q)^2 \frac{F(k, q)^2}{\varepsilon_k + \varepsilon_q + \varepsilon_{k+q}-Q}.
\]

The magnetization becoming negative, Figs. 2 and 3. The second-order corrections remedy this situation. However, close to the boundaries they become very large leading to a very anomalous low field second order magnetization which deviates strongly from the Lanczos results (Fig. 2). Since the deviations between first and second order curves are strongly enhanced close to the classical phase boundaries it is clear that spin wave expansion no longer converges. In Fig. 2 this is obvious for \( \phi/\pi = 0.17 \) at the NAF/CAF boundary and even more for \( \phi/\pi = 0.75 \), already well in advance of the CAF/FM boundary at \( \phi/\pi = 0.85 \). In fact the first order result (long-dashed) agrees better with the Lanczos results (full symbols) plotted for comparison than does the second order curve (short-dashed).

Finally, we mention that from Eq. (29) the second order in \( 1/S \) corrections to the zero-field susceptibility may be calculated in addition to the first order expression given in Eqs. (20) and (22). The comparison of first and second order results for the NAF case is given in Fig. 3 (right panel). Although the second order contributions repair the negative instability of the first order susceptibility at \( \phi \approx 0.11\pi \) the second order result itself diverges when one moves even closer to the classical phase boundary at \( \phi \approx 0.15\pi \).
FIG. 5: Compiled comparison of first (full lines) and second (dotted lines) order in \((1/S)\) spin wave results for the magnetization for various frustration angles indicated in the inset. An offset of 0.2 has been applied. Curves in increasing order correspond to \(\phi\)-values in counter-clockwise direction. Well inside the antiferromagnetic regimes \((\phi/\pi = 0.093, 0.25, 0.65)\) first and second order results show little difference. Close to the classical phase boundaries \((\phi/\pi = 0.135, 0.16, 0.75)\) first order results lead to instable (negative) low-field magnetization. The instability region below \(\phi/\pi = 0.85\) (CAF/FM) is much larger than around \(\phi/\pi = 0.15\) (NAF/CAF). Second order results are always positive but show very anomalous low field magnetization contrary to the Lanczos results in Fig. 2. Note that the second order corrections in \((1/S)\) to the first order curves are positive for low fields and negative for large fields.

IV. EFFECT OF INTERLAYER EXCHANGE COUPLING

In real magnetic systems other interactions may play a certain role besides the in-plane Heisenberg exchange. These include various anisotropies as well as a three-dimensional coupling. Below we shall consider modifications of the above formulas produced by interlayer exchange coupling:

\[
\mathcal{H}_\perp = J_\perp \sum_{\langle ij \rangle_z} \mathbf{S}_i \cdot \mathbf{S}_j ,
\]  

(30)

for nearest-neighbor spins in the direction perpendicular to the layers. (Note the different convention for field direction in Sect. [111]). Here we assume the simple stacking of the layers. The obtained results can be easily extended to other cases as well.
For ferromagnetic exchange $J_\perp < 0$, the saturation field for NAF is still given by $h_s = 8SJ$, while for antiferromagnetic exchange $J_\perp > 0$ it is given by $h_s = 8SJ(1 + \frac{3}{2}J_\perp)$. Here, we define $j_\perp = J_\perp/J_1$. The first-order spin-wave result for the magnetization in the NAF case which includes the interplanar coupling $J_\perp$ is given by

$$J_\perp < 0 : \quad m = S\frac{h}{h_s} \left[ 1 - \frac{1}{2SN} \sum_k \gamma_k \left( \frac{1 + \gamma_k - j(1 - \gamma_k) + \frac{1}{2}j_\perp(1 - \cos k_\perp)}{1 + \gamma_k \cos \theta_c - j(1 - \gamma_k) + \frac{1}{2}j_\perp(1 - \cos k_\perp)} \right)^{1/2} \right],$$

$$J_\perp > 0 : \quad m = S\frac{h}{h_s} \left[ 1 - \frac{1}{2SN} \sum_k \frac{\eta_k}{1 + \frac{1}{2}j_\perp} \left( \frac{1 + \frac{1}{2}j_\perp + \eta_k - j(1 - \gamma_k)}{1 + \frac{1}{2}j_\perp + \eta_k \cos \theta_c - j(1 - \gamma_k)} \right)^{1/2} \right].$$

Here $\eta_k = \gamma_k + \frac{1}{2}j_\perp \cos k_\perp$ is the 3D structure factor and summation is now extended over a 3D Brillouin zone.

In the case of CAF order the first-order spin-wave theory for a ferromagnetic interlayer exchange $J_\perp < 0$ leads to $h_s = 4SJ(1+j)$ and for antiferromagnetic case $J_\perp > 0$ the saturation field is given by $h_s = 4SJ(1+2j+4j_\perp)$. The first order spin wave result for the magnetization in the 3D case is obtained as

$$J_\perp < 0 : \quad m = S\frac{h}{h_s} \left[ 1 - \frac{1}{2SN} \sum_k \frac{j\gamma_k + \frac{1}{2}j_\perp \gamma_k}{j + \frac{1}{2}} \left( \frac{j(1 + \gamma_k) + \gamma_k + \frac{1}{2}j_\perp(1 - \gamma_k)}{j(1 + \gamma_k \cos \theta_c) + \frac{1}{2}(\gamma_k \cos \theta_c \gamma_x) + \frac{1}{2}j_\perp(1 - \gamma_k)} \right)^{1/2} \right],$$

$$J_\perp > 0 : \quad m = S\frac{h}{h_s} \left[ 1 - \frac{1}{2SN} \sum_k \frac{j\gamma_k + \frac{1}{2}j_\perp \gamma_k + \frac{1}{2}j_\perp \gamma_x}{j + \frac{1}{2}(1 + j_\perp)} \left( \frac{j(1 + \gamma_k) + \gamma_k + \frac{1}{2}j_\perp (1 + \gamma_k)}{j(1 + \gamma_k \cos \theta_c) + \frac{1}{2}(\gamma_k \cos \theta_c \gamma_x) + \frac{1}{2}j_\perp (1 + \gamma_k \cos \theta_c)} \right)^{1/2} \right].$$

Here we used the convention $\gamma_i = \cos k_i$ ($i = x, y, z$).

We have verified that for well ordered NAF and CAF phases the effect of interlayer coupling is hardly visible up to $J_\perp \sim 0.3J_{1,2}$. This means that the application of our first order spin wave results to real compounds does not critically depend on a small interlayer coupling $J_\perp$ as long as the quantum antiferromagnet is in a well-ordered phase. Closer to the phase boundaries $J_\perp$ has the the effect of stabilizing the ordered phases, especially for $J_\perp < 0$.

V. DISCUSSION AND CONCLUSION

In this work we have explored the quantum corrections to the magnetization, the uniform susceptibility and the canting angle in the first- and the second-order spin-wave approximation and by using exact diagonalization of finite clusters. The deviations from classical behaviour were found to be pronounced close to the strongly frustrated regions where the classical phases meet. Indeed we have shown that linear spin-wave theory breaks down in these regions and second-order corrections do not fundamentally change this observation. The latter are small well within the AF regions and have positive sign for small and negative sign for large fields. Although they prevent the instability of the linear spin-wave theory for small fields they become very anomalous close to the boundaries and one cannot expect that the $1/S$ expansion converges in the nonmagnetic region. This is intuitively clear since the spin wave expansion in the region of the classical boundaries starts from the wrong, i.e., magnetically ordered ground state.

A striking feature of these results is that the deviations from classical behaviour in the magnetization curve $m(h)$ and the breakdown of the spin-wave expansion are most pronounced on the ferromagnetic side ($J_1 < 0$), as illustrated in Figs. 2 and 5. One can understand this by considering the asymptotic form of the first-order corrections to the staggered moments $\delta S$ and the uniform susceptibility $\delta \chi$ in the CAF phase as $j \to \pm1/2$:

$$\delta S \approx \sum_k \frac{1}{k^2_x + (\delta j|k^2_y)},$$

$$\delta \chi \approx \frac{1}{j + \frac{1}{2}} \sum_k \frac{1}{k^2_x + (\delta j|k^2_y)}. \quad (33)$$

Here, $\delta j = j \pm \frac{1}{2}$ is the deviation from one of the two strongly frustrated points. For the CAF/NAF boundary the diverging corrections are the same for the sublattice magnetization and the susceptibility: $\delta S, \delta \chi \sim \ln |\delta j|$. 

11
finite clusters is observed. The classical value of the frustration angle \( \phi \) where the magnetization is plotted as function of the wave theory lends further support to this idea.

The pronounced quantum fluctuations seen in spin wave results according to Eq. (15) with respect to the constant classical value \( \mu / \mu_B = m / S = 0.58 \). At the FM/NAF boundary where the quantum fluctuations are small the magnetization starts at this value and then is continuously reduced as one approaches the strongly frustrated sector around NAF/CAF boundary. There the magnetization jumps to a slightly larger value and then breaks down close to the spin nematic region around CAF/FM boundary. Right: Susceptibility \( \chi(\phi) / \chi(\phi = 0) \) for \( h = h_c(\phi) \) normalized to the simple NAF value. Circles are ED Lanczos results (\( k_B T/J_c = 0.2 \)) and full lines are \( T=0 \) spin wave results according to Eq. (20).

For the CAF/FM frustration point the susceptibility correction acquires an additional diverging prefactor: \( \delta \chi \sim (\ln |\delta j|) / |\delta j| \), which indicates that a long-range magnetic order is destabilized in a much wider window of \( J_2 \) for this sign of \( J_1 \). This is in accordance with the ED Lanczos results where a tendency to bound state formation of spin waves as indicated by the \( \Delta S_z = 2 \) steps in the magnetization of finite clusters is observed. This is evidence that around \( \phi \approx 0.85 \pi \) or \( J_2 / J_1 \approx -0.5 \) the ground state will be of the spin nematic type as proposed in Ref. [17]. It may be described as a quantum gapless phase with a Goldstone mode describing the collective long range excitations of a nonlocal quadrupolar order parameter. A second order transition between and the CAF phase and this spin nematic is permitted by the symmetry of the order parameter, and the smooth evolution of ED spectra across the transition suggest that a second order does in fact occur. The pronounced quantum fluctuations seen in spin wave theory lend further support to this idea.

This can be most clearly seen in Fig. 6 (left panel) where the magnetization is plotted as function of the frustration angle \( \phi \). The decrease in magnetization from the classical value \( m / S = h / h_c \) at around half saturation \( h / h_c = 0.58 \) characterizes the strength of quantum fluctuations. Their effect increases from zero at FM/NAF boundary to a maximum at NAF/CAF boundary where a discontinuous jump in the magnetization occurs. For \( \phi / \pi > 0.5 \) (the CAF regime with \( J_2 < 0 \)) the reduction of \( m / S \) due to quantum fluctuations rapidly becomes large and the expansion in \( 1 / S \) breaks down. This is also seen in Fig. 5.

The exact numerical results (circles) obtained from the Bonner-Fisher plots (hence the steps) as in Fig. 4 also show the strong reduction of the magnetization close to classical CAF/FM boundary. This is a signature of the true spin-nematic quantum ground state which does not break time reversal symmetry. Therefore it has no first order (linear) coupling to the magnetic field, resulting in a small magnetization. The right panel of Fig. 6 shows the comparison of high-field susceptibility at \( h = h_c(\phi) \) from first order spin wave theory (full line) at \( T=0 \) with the ED Lanczos results at small but finite \( T \), plotted as function of the frustration angle \( \phi \). The susceptibility in both cases is normalized to the pure NAF case \( (\phi = 0) \). The overall agreement of \( \phi \) dependence is quite good because due to high fields the effect of quantum fluctuations is suppressed. Again the deviations are strongest close to the classical CAF/FM boundary where the saturation field approaches zero and the spin wave approximation breaks down.

The deviations from classical results and the breakdown of spin wave approximation is much less severe on the CAF/NAF phase boundary at the AF side \( (J_1 > 0) \). The magnetization behaves significantly less singular in this region. As mentioned before a discontinuous jump in \( m(\phi) \) at \( \phi \approx 0.15 \pi \) occurs around half saturation (Fig. 5). Then the quantum phase transition to the presumably stacked spin dimer ground state may be expected to be a first order transition. Therefore the magnetic and non-magnetic phases on both sides of the boundary will correspond to stable local minima of the free energy and
fluctuations will not be very pronounced leading to a less singular magnetization behaviour. The first-order nature of transition between CAF and a nonmagnetic columnar dimer state was noticed in early numerical work while for the boundary with the NAF, a first-order scenario was put forward only recently.

Finally, we comment on experimental data for the high field magnetization of $J_1$–$J_2$ compounds. There are no published data for those systems mentioned in the introduction. However recently a new compound (CuBr)LaNb$_2$O$_7$, with a perovskite/metal halide intergrowth structure was synthesized. This is a spin-1/2 magnetic insulator, and is reported to exhibit quasi-2D magnetic behavior arising from the CuBr - square lattice planes. It shows CAF order at a relatively large $T_N = 32$ K.

It has been suggested that the magnetism of (CuBr)LaNb$_2$O$_7$ can be described by the square lattice $J_1$–$J_2$ model with a frustration angle $\phi = 0.73\pi$ ($J_2/J_1 = -1.1$), which puts this compound closer to the strongly frustrated region of the spin nematic phase than any other compound reported so far. Our spin wave calculations predict a pronounced nonlinear magnetization for this parameter set, particularly at low fields. However, the experimental results in Ref. show only a modest curvature, at high fields. We therefore conclude that, while these materials likely do possess competing FM and AF interactions, they probably cannot be described by a simple square lattice $J_1$–$J_2$ model.

In conclusion, it is abundantly clear that classically disordered phases of the square lattice $J_1$–$J_2$ model need and deserve an analysis which goes beyond the spin wave theory presented here. For further progress to be made in understanding the “hidden order” phases an approximate treatment of the broken order parameter and its low lying excitations is necessary. Since both the dimer and nematic phases are bond centered, the bond-operator method might be a useful choice.

VI. ACKNOWLEDGEMENT

We are pleased to acknowledge helpful conversations with C. Geibel, Tsutomu Momoi and Luis Seabra. This work was supported in part by the SFB 463 project of DFG and by EPSRC Grant (No. EP/C539974/1).

1 G. Misguich and C. Lhuillier. in Frustrated Spin Systems ed. by H. T. Diep (World Scientific, Singapore, 2004)
2 A. Laeuchli, J.C. Domenges, C. Lhuillier, P. Sindzingre and M. Troyer, Phys. Rev. Lett. 95, 137206 (2005)
3 G. Misguich and P. Sindzingre, [arXiv:0704.1017v2, to appear in Eur. Phys J. B]
4 P. Millet and C. Satto, Mat. Res. Bull. 33, 1339 (1998).
5 R. Melzi, P. Carretta, A. Lascialfari, M. Mambrini, M. Troyer, P. Millet, and F. Mila, Phys. Rev. Lett. 85, 1318 (2000).
6 R. Melzi, S. Aldrovandi, F. Teobaldi, P. Carretta, P. Millet, and P. Mila, Phys. Rev. B 64, 024409 (2001).
7 E. E. Kaul, H. Rosner, N. Shannon, R. V. Shapchenko and C. Geibel, J. Magn. Magn. Mater. 272-276, 922 (2004)
8 E. Kaul, PhD thesis, Technische Universität Dresden (2005)
9 N. Kini, E. E. Kaul and C. Geibel, Journal of Physics: Condensed Matter 18, 1303 (2006).
10 N. Shannon, B. Schmidt, K. Penc, and P. Thalmeier, European Physical Journal B 38, 599 (2004).
11 B. Schmidt, P. Thalmeier and N. Shannon, Phys. Rev. B 76, 125113 (2007).
12 M. P. Gelfand, R. R. Singh and D. A. Huse, Phys. Rev. B 40, 10801 (1989).
13 S. Sachdev and R. N. Bhatt, Phys. Rev. B 41, 9323 (1990).
14 H. J. Schulz and T. A. L. Ziman, Europhys. Lett. 18, 355 (1992).
15 V. N. Kotov, J. Oitmaa, O.P. Sushkov and Zheng Weihong, Phys. Rev. B 60, 14613 (1999).
16 J. Sirker, Z. Weihong, O.P. Sushkov and J. Oitmaa Phys. Rev. B 73, 184420 (2007).
17 N. Shannon, T. Momoi, and P. Sindzingre, Phys. Rev. Lett. 96, 027213 (2006).
18 M. E. Zhitomirsky and T. Nikuni, Phys. Rev. B 57, 5013 (1998).
19 M. S. Yang and K. Mütter, Z. Phys. B 104, 117 (1997).
20 J. C. Bonner and M. S. Fisher, Phys. Rev. 135, A640 (1964).
21 S. Gluzman, Z. Phys. B 90, 313 (1993).
22 M. E. Zhitomirsky, A. Honecker, and O. A. Petrenko, Phys. Rev. Lett. 85, 3269 (2000).
23 A. Honecker, Can. J. Phys. 79, 1557 (2001).
24 G. Jackeli and M. E. Zhitomirsky, Phys. Rev. Lett. 93, 017201 (2004).
25 N. Oba, H. Kageyama, T. Kitano, J. Yasuda, Y. Baba, M. Nishii, K. Hirota, Y. Narumi, M. Hagiwara, K. Kindo, T. Saito, Y. Ajiro and K. Yoshimura, J. Phys. Soc. Jpn. 75, 113601 (2006).