Enhancement of Anisotropy Due to Fluctuations in Quasi-One-Dimensional Antiferromagnets

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Abstract

It is shown that the observed anisotropy of magnetization at high magnetic fields in RbMnBr$_3$, a quasi-one-dimensional antiferromagnet on a distorted stacked triangular lattice, is due to quantum and thermal fluctuations. These fluctuations are taken into account in the framework of linear spin-wave theory in the region of strong magnetic fields. In this region the divergent one-dimensional integrals are cut off by magnetic field and the bare easy-plane anisotropy. Logarithmical dependence on the cutoff leads to the "enhancement" of the anisotropy in magnetization. Comparison between magnetization data and our theory with parameters obtained from neutron scattering experiments has been done.

I. INTRODUCTION

The question of the effect of fluctuations on the magnetic properties of quasi-one-dimensional antiferromagnets has been intensively discussed in the last few years. Particular
attention has been given to the cases where there is a stacked triangular lattice and an antiferromagnetic interaction, so that there is a frustration in going from one-dimensional to three-dimensional ordering.

This situation is apparent in materials with hexagonal CsNiCl$_3$-type crystal structure having the general formula ABX$_3$ where A is an alkali metal, B is a bivalent metal of 3d group, and X is a halogen. While spin dynamics of CsNiCl$_3$ and RbNiCl$_3$ (S=1, easy-axis anisotropy) has been studied to examine Haldane conjecture [1,2], the compounds of the same group, CsMnBr$_3$, RbMnBr$_3$ (S=5/2, easy-plane anisotropy) can be considered as appropriate for checking the applicability of standard spin-wave theory for the case of large half-integer spins.

CsMnBr$_3$ has been investigated by various experimental techniques as an example of a frustrated antiferromagnet on a stacked triangular lattice exhibiting a number of unusual magnetic properties, such as a field-induced phase transition from the triangular phase to the collinear one, critical behavior associated with a chiral degeneracy and an unusual phase diagram. In addition to numerous neutron scattering experiments [3] and to ESR measurements [4], the magnetization process has been studied in detail [5–8] with results which are in good agreement with classical calculations [5] except for two discrepancies. Namely:

- the measured magnetic torques are significantly smaller than the theoretical ones;
- in large magnetic fields ($H > H_c$, where $H_c$ is the critical field of the transition to the collinear phase) there is a considerable anisotropy between magnetization when magnetic field is applied along C-axis of the crystal ($M^{H\parallel C}$) and the one ($M^{H\perp C}$) for magnetic field along easy plane absent in the theory [5].

It was suggested [8] that in order to describe these peculiarities the anisotropy of quantum fluctuations should be taken into account. In this paper we consider on the basis of another compound RbMnBr$_3$ how both quantum and thermal fluctuations affect the magnetization of quasi-one-dimensional noncollinear antiferromagnet.
II. CRYSTAL STRUCTURE AND MAGNETIC PROPERTIES OF RbMnBr$_3$

Powder neutron scattering measurements by Glinka et al. [9] showed that RbMnBr$_3$ exhibits antiferromagnetic order below $T_N = 8.8 \pm 0.1$K. Single crystal measurements [10,11] show that the magnetic structure of RbMnBr$_3$ is incommensurate with $Mn^{+2}$ moments lying in the basal plane and with antiferromagnetic ordering in the $c$ direction. The commensurate ordering in CsMnBr$_3$ gives scattering peaks of type $(\frac{1}{3}, \frac{1}{3}, 1)$ [3] while in the incommensurate RbMnBr$_3$ these peaks are replaced by two triads of peaks near to $(\frac{1}{3}, \frac{1}{3}, 1)$ with three fold symmetry around $(\frac{4}{3}, \frac{4}{3}, 1)$. Because the incommensurate peaks surround a commensurate position in reciprocal space, it is likely that the incommensurate structure is locally similar to the 120° triangular structure of CsMnBr$_3$, but with angles between neighboring spins in the basal plane somewhat larger than 120°.

The appearance of incommensurate magnetic structure may be due to the presence of crystal structure distortions. According to X-ray study by Von Fink and Seifert [12] as the temperature decreases the crystal structure of RbMnBr$_3$ changes at 470K from CsNiCl$_3$-type structure (space group $P6_3/mmc$) to the so-called KNiCl$_3$-type structure (space group $P6_3cm$). Measurements of the birefringence [13] and recent neutron scattering experiments [14] show that another structural phase transition takes place at about 220K. As reported in [14] below this temperature the crystal has orthorhombic structure with cell dimensions $a=14.680\ \text{Å}$, $b=12.805\ \text{Å}$, $c=6.516\ \text{Å}$ at 12K.

At low temperatures a field of 3.0T applied along the $a$ axis produces a first-order phase transition to a commensurate structure [11,13], so that the lattice parameter of the magnetic cell is eight times that of the nuclear cell [10]. In a magnetic field of about $H_c = 3.9$T and above applied in the basal plane, the magnetic structure becomes collinear, as in the case of CsMnBr$_3$ with $H_c = 6.4$T. This conclusion can be derived from a previous study of the magnetization process [16] and ESR-spectrum [17] in RbMnBr$_3$.

The crystal distortions lead primarily to a change in the antiferromagnet coupling between chains. But for the purpose of this paper the small distinction between the RbMnBr$_3$
magnetic structure and a simple 120°-structure is not so important. Moreover confining ourselves by the range of large magnetic fields we assume that the effects of inter-chain exchanges and particularly the distortion of a simple 120° magnetic structure in the absence of magnetic fields will give only quantitative corrections not changing the qualitative picture. This assumption appears to be true and we estimate these corrections for large magnetic fields (see [19-23] below).

III. EXPERIMENTAL PROCEDURE AND RESULTS

A vibration-sample magnetometer, similar to the one described in ref. [18], was used to measure the magnetization in RbMnBr$_3$. A magnetic field up to 6T was generated by two superconducting coils. Simultaneous measurements of two mutually perpendicular components of the magnetization of the sample, one of which ($M_{\text{long}}$) is parallel and another ($M_{\text{tr}}$) is perpendicular to the direction of magnetic field were done by using three pairs of measuring coils.

The absolute accuracy of the magnetization measurements was about 5%. The crystal was oriented with an accuracy of 1-2°.

The investigation was performed on single crystals having approximately $1.5 \times 1.5 \times 1.5 mm^3$ volume and approximately 30mg mass in the temperature range 1.7-12K.

The magnetization component $M_{\text{long}}$ parallel to the field as a function of the field $H$ for $H\parallel C$ and $H\perp C$ at $T=1.7K$ is shown in Fig.1. In fields above the critical field ($H_c \approx 3.9T$) the magnetizations $M_{\text{long}}^{H\parallel C}$ and $M_{\text{long}}^{H\perp C}$ are not the same, but differ by about 7%. This anisotropy of magnetization can not be explained just by taking into account the easy plane anisotropy in classical calculations, because it leads to $M_{\text{long}}^{H\parallel C} < M_{\text{long}}^{H\perp C}$ while experimentally the situation is opposite. Moreover corrections to the classical magnetization due to easy-plane anisotropy have to be of order $D/J \approx 1\%$ which is much smaller than the observed anisotropy ($5 - 10\%$).

The anisotropy in magnetization is shown on Fig.2 where the magnetization component
$M_{tr}$ perpendicular to the magnetic field $H$ as a function of the field is plotted at different angles $\varphi$ between the field and the basal plane and at $T=4.2$K. Due to the anisotropy of the magnetization at $H > H_c$, a non zero transverse-magnetization signal occurs at $\varphi \neq 0$.

According to Fig.2 the field dependence $M_{tr}(H)$ exhibits a small hysteresis. This hysteresis takes place when the projection of the magnetic field on the basal plane reaches the value of $3.19 \pm 0.10$T in increasing fields and $2.67 \pm 0.07$T in decreasing fields. It corresponds to the first order phase transition from the incommensurate magnetic structure to the commensurate one.

In Fig.3 the field dependence of the magnetization component $M_{tr}$ at different temperatures is shown. At large magnetic fields we can see that $M_{tr}$ increases with temperature supporting the idea that the anisotropy of the magnetization at $H > H_c$ is due to fluctuations.

This is even clearer from Fig.4 where the value of $M_{tr}(H = 4.5$T, $\varphi = 20^\circ)$ is shown as a function of temperature.

IV. THEORY

We start with the microscopic Hamiltonian

$$H = 2J \sum_i \vec{S}_i \vec{S}_{i+\Delta_z} + 2J' \sum_i \vec{S}_i \vec{S}_{i+\Delta_\perp} + D \sum_i (S^z_i)^2 - \vec{h} \sum_i \vec{S}_i$$

(1)

The intra-chain antiferromagnet exchange constant $J$ is the largest parameter in the Hamiltonian (1) and is about two orders of magnitude larger than the exchange constant between chains $J'$ and easy-plane single-ion anisotropy $D$ [11], $J >> J'$, $D$ and $J, J', D > 0$. $\vec{h}$ is the magnetic field in the units of energy and it is equal to $h = g\mu_B H$ where $H$ is a magnetic field in some standard units, $\mu_B$ is a Bohr magneton and $g$ is Lande factor which is equal 2 in the compound under consideration. For $h = 0$ the classical ground state of the Hamiltonian (1) is the 120° structure with spin vectors forming equilateral triangles in the basal plane. The case of a non zero magnetic field was considered classically by Chubukov [11]. For $\vec{h}$ along the
Z (C) axis the transverse components of the spins conserve 120° structure and the classical magnetization

\[ M_{h\parallel C} \equiv \langle S^z \rangle = \frac{h}{8J} \quad (2) \]

when \( h < 8JS \) neglecting by \( J'/J, D/J \) (here and there on \( M \) means \( M_{\text{long}} \) where the opposite is not stated explicitly). The case of the magnetic field perpendicular to the Z axis is more complicated. For a sufficiently strong easy-plane anisotropy, spins do not leave the planes but the 120° structure is not conserved anymore and a spin flip of the two magnetic sublattices takes place at the relatively small magnetic field \( h_c = \sqrt{48JJ' S^2} \). At \( h_c < h < 8JS \) we have a collinear spin structure and magnetization has to be the same (up to small \( J', D/J \) corrections) as in the case of field along Z axis [5].

In this section we take into account quantum and thermal fluctuations in the framework of linear spin-wave theory and show that the anisotropy of magnetization at \( h > h_c \) due to these fluctuations is of the right sign (\( M_{h\perp C} < M_{h\parallel C} \)) and is much larger than the one expected from classical calculation.

In what follows we confine ourselves to magnetic fields \( h > h_c \) where we have a collinear spin structure for a magnetic field perpendicular to the C axis of the crystal and we neglect the \( J' \) term in the Hamiltonian ([1]) in the body of this chapter. This term will give corrections to the gap in a spin-wave spectrum of order of \( J'/J \) without changing the qualitative picture (see (21-22) below). Thus we consider the Hamiltonian of a single magnetic chain instead of ([1]) keeping in mind that our results will be applicable only for the \( h > h_c = \sqrt{48JJ' S^2} \) region. We can write Hamiltonians for two directions of magnetic field in the form:

\[ H_{h\parallel C} = 2J \sum_i \vec{S}_i \cdot \vec{S}_{i+1} - h \sum_i S^z_i + D \sum_i (S^z_i)^2 \quad (3) \]

\[ H_{h\perp C} = 2J \sum_i \vec{S}_i \cdot \vec{S}_{i+1} - h \sum_i S^z_i + D \sum_i (S^x_i)^2 \quad (4) \]

Here we slightly changed notations for simplicity. Now magnetic field is always applied along Z axis and the easy plane is XY plane for the parallel case and YZ plane for the perpendicular one. Let us apply usual Dyson-Maleev transformation
\[ S' = -S + a^+a \]
\[ S^+ = \sqrt{2S}a^+(1 - \frac{a^+a}{2S}) \]
\[ S^- = \sqrt{2S}a \]  

(5)

to (3,4) with the axes of quantization \( z' \) directed as it is shown in Fig. 5. Choosing the angle \( \phi \) to cancel linear in \( a, a^+ \) terms in Hamiltonians (3,4) we reproduce the classical results for magnetization. The terms of (3,4) quadratic in \( a, a^+ \) will give us the linear spin-wave theory corrections to these results. We have:

\[
\sin \phi_\parallel = -\frac{h}{2S(4J + D)} \\
\sin \phi_\perp = -\frac{h}{8JS} 
\]

(6)

(7)

and both Hamiltonians (3,4) after Fourie transformation can be written in the form

\[
\mathcal{H} = \gamma + \sum_k \left[ \omega_1(k)a^+_ka_k - \frac{\omega_2(k)}{2}(a^+_ka_{-k} + a_ka_{-k}) \right] 
\]

(8)

with

\[
\gamma_\parallel = -N(2JS^2(1 + 2\sin^2 \phi) - \frac{DS}{2}\cos^2 \phi + DS^2\sin^2 \phi) \\
\omega_{1\parallel}(k) = 4JS(1 - \sin^2 \phi \cos k) + DS\cos^2 \phi \\
\omega_{2\parallel}(k) = 4JS\cos^2 \phi \cos k + DS\cos^2 \phi \\
\gamma_\perp = -N(2JS^2(1 + 2\sin^2 \phi) - \frac{DS}{2} \\
\omega_{1\perp}(k) = 4JS(1 - \sin^2 \phi \cos k) + DS \\
\omega_{2\perp}(k) = 4JS\cos^2 \phi \cos k - DS
\]

Making Bogolyubov transformation

\[
a_k = \left( \frac{\omega_1 + \sqrt{\omega_1^2 - \omega_2^2}}{2\sqrt{\omega_1^2 - \omega_2^2}} \right)^{1/2} b_k + \left( \frac{\omega_1 - \sqrt{\omega_1^2 - \omega_2^2}}{2\sqrt{\omega_1^2 - \omega_2^2}} \right)^{1/2} b_{-k} 
\]

(9)

we diagonalize (8) and get

\[
\mathcal{H} = \gamma + \sum_k \left[ \epsilon(k)(b_k^+b_k + \frac{1}{2}) - \frac{\omega_1(k)}{2} \right] 
\]

(10)
with the spectrum of spin waves given by

$$\epsilon^2(k) = \omega_1^2(k) - \omega_2^2(k)$$

or

$$\epsilon_\parallel^2(k) = 4JS(1 - \cos k) \left[ 4JS(1 + \cos 2\phi \cos k) + 2DS \cos^2 \phi \right]$$

(11)

$$\epsilon_\perp^2(k) = [4JS(1 - \cos k) + 2DS] 4JS(1 + \cos 2\phi \cos k)$$

(12)

Now we can find the magnetization using general relation $M = -< \frac{\partial H}{\partial h} >$

$$M = -\frac{\partial \gamma}{\partial h} + \sum_k \left[ -\frac{\partial \epsilon(k)}{\partial h} (<b^*_k b_k> + \frac{1}{2}) + \frac{1}{2} \frac{\partial \omega_1}{\partial h} \right]$$

(13)

or after simple manipulations using $<b^*_k b_k> + \frac{1}{2} = \coth \frac{\epsilon}{2T}$

$$M = -S \sin \phi - \int_{-\pi}^{\pi} \frac{dk}{2\pi} \left[ -\frac{\partial \epsilon(k)}{\partial h} \right] \frac{1}{2} \coth \frac{\epsilon}{2T}$$

(14)

Here the first term describes the classical part of magnetization while the second one comes from the contribution of quantum and thermal fluctuations.

Let us show now that Eq. (14) leads to the enhanced anisotropy between $M_{h\parallel C}$ and $M_{h\perp C}$. Consider the case of zero temperature for simplicity. Substituting 1 for $\coth \frac{\epsilon}{2T}$ and taking derivatives using (6, 7, 11, 12) we have

$$M_{h\parallel C} = -S \sin \phi$$

$$-\sin \phi \int_{-\pi}^{\pi} \frac{dk}{2\pi} \frac{1}{2} \sqrt{\frac{(1 - \cos k)}{1 + \cos 2\phi \cos k + D/2J \cos^2 \phi}} \frac{\cos k + D/4J}{1 + D/4J}$$

$$M_{h\perp C} = -S \sin \phi - \sin \phi \int_{-\pi}^{\pi} \frac{dk}{2\pi} \frac{1}{2} \sqrt{\frac{1 - \cos k + D/2J}{1 + \cos 2\phi \cos k}} \cos k$$

(15)

Now it is clear that only one of two soft modes of spin waves corresponding $k = \pi$ gives a big contribution to the magnetization. This result is quite obvious because it is this mode that corresponds to fluctuations of the angle between spins and magnetic field. Another soft mode at $k = 0$ corresponds to azimuthal fluctuations of spins around the direction of magnetic field and almost does not contribute to magnetization. This mode however contributes very
much to the value of average spin on the site. Expanding the integrands in Eq.(15) in the vicinity of \( k = \pi \) and neglecting \( D/J \) in comparison with 1 we get

\[
M \approx -S \sin \phi + \sin \phi \int \frac{dk}{2\pi} \frac{1}{\sqrt{k^2 + g^2}}
\]

with

\[
g_\parallel^2 = \frac{h^2}{16J^2S^2} + \frac{D}{J}
\]

\[
g_\perp^2 = \frac{h^2}{16J^2S^2}
\]

or taking integrals finally

\[
M_{h\parallel C} \approx \frac{h}{8J} \left( 1 + \frac{1}{2\pi S} \ln \left( \frac{h^2}{16J^2S^2} + \frac{D}{J} \right) \right)
\]

\[
M_{h\perp C} \approx \frac{h}{8J} \left( 1 + \frac{1}{2\pi S} \ln \left( \frac{h^2}{16J^2S^2} \right) \right)
\]

Now let us estimate the corrections to (19-20) due to 3D \((J')\) effects. These effects give corrections of order of \( J'/J \) under logarithm in (19-20) and can be calculated in the framework of the same linear spin wave theory. Calculations show that 3D fluctuations will effectively increase the gap of 1D system giving instead of (19-20):

\[
M_{h\parallel C} = \frac{h}{8J} \left[ 1 + \frac{1}{2\pi S} \ln \left( \frac{h^2}{16J^2S^2} + \frac{D}{J} + 3\frac{J'}{J} \right) \right]
\]

\[
M_{h\perp C} = \frac{h}{8J} \left[ 1 + \frac{1}{2\pi S} \ln \left( \frac{h^2}{16J^2S^2} + 2\frac{J'}{J} \right) \right]
\]

The formulas (21, 22) are good at sufficiently big magnetic fields where fluctuations and effects of three-dimensional ordering are small enough to guarantee the applicability of linear spin wave theory for quasi-one-dimensional system.

\footnote{It is seen from this argument that the attempts \cite{19} to relate the suppression of magnetization with the suppression of average spin by fluctuations are qualitatively wrong because these effects arise from the different modes of spin waves.}

\footnote{Expressions like \( \ln \frac{J'}{J} \) were obtained in \cite{20} for spin reduction but can not be applied to our case (see footnote 1).}
Now from (19-22) the origin of the enhancement of anisotropy is clear. Big fluctuations of the angle between spins and magnetic field give a significant contribution to magnetization which depends strongly on the corresponding gap in the spectrum of spin waves due to low dimensionality of the system (integral in Eq. (16) would diverge for \( g = 0 \)). This gap is larger for the case of magnetic field along \( C \)-axis due to bare easy-plane anisotropy in (1).

V. COMPARISON WITH EXPERIMENT AND DISCUSSION

To compare our theory with an experiment we need to know the values of the parameters of the Hamiltonian (1) obtained from some independent source. The inelastic neutron scattering experiments provide us with such a source. As reported in [10] \( \tilde{J} = 199 \text{GHz} \) and \( \tilde{D} = 2.2 \text{GHz} \). These values were found as the best ones to fit the classical formulas for spin-wave dispersion to the data of an inelastic neutron scattering. To get the values of \( J \) and \( D \) in (1) we have to take into account the renormalizations by quantum fluctuations. Using (4) and (5) from [5] we have:

\[
J = 186 \text{GHz}, D = 1.3 \text{GHz}
\]

Using these values of \( J \) and \( D \) and the expressions (21) and (22) with \( J' = 0.22 \text{GHz} \) from \( H_c = \sqrt{48JJ'S^2} = 3.9 \text{T} \) we have calculated the magnetization of RbMnBr\(_3\) for both \( H \parallel C \) and \( H \perp C \). The range of applicability of (21) and (22) is approximately \( H > 4 \text{T} \). For \( H < 4 \text{T} \) the effects of three dimensional ordering and next order corrections to the linear spin-wave theory have to be seriously taken into account.

The magnetizations (21-22) are plotted in Fig.1 together with the experimental data for \( M_{H\parallel C} \) and \( M_{H\perp C} \). Also we plotted the magnetizations calculated using classical formulas and the values \( J = 199 \text{GHz}, D = 2.2 \text{GHz} \). We can see that the formulas (21) and (22) give both the much better absolute values of the magnetizations then the classical expressions and describe the anisotropy in magnetization. One can notice also that the experimental dependence \( M_{H\parallel C} \) is a little bit nonlinear with the slope increasing with \( H \) (this is not clearly
seen from Fig.1 but can be checked by fitting the experimental data by linear functions in different ranges of $H$). This can be naturally explained by suppressing fluctuations with increasing $H$ and it is seen in (21).

The graphs obtained from (21-22) in Fig.1 lie below experimental points. This probably can be corrected by the second order in $1/S$ corrections to magnetization which will contribute with the sign opposite to the sign of the first order corrections. The contribution of the higher order terms in $1/S$ is not very small because the fluctuations are quite big in the experimental range of parameters. For example $\frac{\Delta M_{H<0}}{M_{\text{class}}} \approx 30\%$ at $H = 4.5T$.

VI. CONCLUSION

The new effect of enhancement of anisotropy in quasi-one-dimensional spin systems due to quantum and thermal fluctuations was found. It explains the anisotropy in magnetization seen in experiments. The essence of the effect is that the fluctuational part of magnetization is sufficiently big and is determined mostly by logarithm of the gap in the spin wave spectrum. This gap is anisotropic due to the bare easy-plane anisotropy. The logarithmical dependence of the fluctuational part of the magnetization on this gap (essentially the divergence of one-dimensional fluctuations) leads to the strong ”enhanced” anisotropy of the magnetization. The same fluctuations explain a slightly nonlinear character of $M_{H \parallel C}$ dependence and the excessive values of magnetization calculated using classical formulas.

This theory can be applied to the compounds with easy plane anisotropy and quasi-one-dimensional magnetic structure such as RbMnBr$_3$, CsMnBr$_3$ and KNiCl$_3$.

\footnote{For the magnetization data for CsMnBr$_3$ taken from [7,8] formulas (21-22) give even the better fit then for RbMnBr$_3$ in Fig.1.}
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**FIGURE CAPTIONS**

**Figure 1** The magnetization $M_{\text{long}}$ parallel to the direction of magnetic field as a function of field. Circles (○) denote $M_{\parallel}(H)$ for $H \parallel C$ and squares (□) denote $M_{\perp}(H)$ for $H \perp C$. Solid and dashed lines are the results of calculation with parameters $J = 186\text{GHz}$, $D = 1.3\text{GHz}$, $H_c = 3.9\text{T}$, $J' = 0.22\text{GHz}$ using formulas (21-22) and classical theory [5] with $J = 199\text{GHz}$, $D = 2.2\text{GHz}$ respectively.

**Figure 2** The magnetization $M_{\text{tr}}$ perpendicular to the magnetic field as a function of $H$ with the field making small angles $\varphi$ with the basal plane. $\varphi = 36^\circ$ (1), $\varphi = 26^\circ$ (2), $\varphi = 16^\circ$ (3), $\varphi = 0.5^\circ$ (4), $\varphi = -14^\circ$ (5), $\varphi = -24^\circ$ (6), $\varphi = -34^\circ$ (7).

**Figure 3** The field dependence of the magnetization component $M_{\text{tr}}$ at different temperatures with the field making angle $\varphi = 20^\circ$ with the basal plane. $T = 2.4\text{K}$ (1), $T = 4.2\text{K}$ (2), $T = 6.0\text{K}$ (3), $T = 7.1\text{K}$ (4).

**Figure 4** The magnetization $M_{\text{tr}}(H = 4.5\text{T}, \varphi = 20^\circ)$ is shown as a function of temperature. The dotted line is a guide to an eye.

**Figure 5** Chosen quantization axes and coordinate systems.