BaV$_3$O$_8$: A possible Majumdar-Ghosh system with $S=1/2$

T. Chakrabarty, A. V. Mahajan, A. A. Gippius, A. V. Tkachev, N. Büttgen, and W. Kraetschmer

1Department of Physics, IIT Bombay, Powai, Mumbai 400076, India
2Faculty of Physics, Moscow State University, Moscow 119991, Russia,
A.V. Shubnikov Institute of Crystallography, Moscow 119333, Russia
3Institute of Physics, University of Augsburg, D-86135 Augsburg, Germany

Abstract

BaV$_3$O$_8$ contains magnetic V$^{4+}$ ($S=1/2$) ions and also non-magnetic V$^{5+}$ ($S=0$) ions. The V$^{4+}$ ions are arranged in a coupled Majumdar-Ghosh chain like network. A Curie-Weiss fit of our magnetic susceptibility $\chi(T)$ data in the temperature region of 80 – 300K yields a Curie constant $C = 0.39$ cm$^3$/K/mol V$^{4+}$ and an antiferromagnetic Weiss temperature $\theta = -26$K. The $\chi(T)$ curve shows a broad maximum at $T \sim 25$K indicative of short-range order (SRO) and an anomaly corresponding to long-range order (LRO) at $T_N \sim 6K$. The value of the ‘frustration parameter’ $(f = |\theta/T_N| \sim 5)$ suggests that the system is moderately frustrated. Above the LRO temperature the experimental magnetic susceptibility data match well with the coupled Majumdar-Ghosh (or $J_{nn} - J_{nnn}$ Heisenberg) chain model with the ratio of the $nnn$ (next-nearest neighbor) to $nn$ (nearest neighbor) magnetic coupling $|z| = 2$ and $J_{nnn}/k_B = 40$K. In a mean-field approach when considering the inter-chain interactions, we obtain the total inter-chain coupling to be about 16K. The LRO anomaly at $T_N$ is also observed in the specific heat $C_v(T)$ and is not sensitive to an applied magnetic field up to 90 kOe. A $^{51}$V NMR signal corresponding to the non-magnetic vanadium was observed. Anomalies at 6K were observed in the variation with temperature of the $^{51}$V NMR linewidth and the spin-lattice relaxation rate $1/T_1$ indicating that they are sensitive to the LRO onset and fluctuations at the moment V sites. The existence of two components (one short and another long) is observed in the spin-spin relaxation rate $1/T_2$ data in the vicinity of $T_N$. The shorter component seems to be intimately connected with the magnetically ordered state. We suggest that both magnetically ordered and non-long range ordered (non-LRO) regions coexist in this compound below the long range ordering temperature.

PACS numbers: 75.10.Pq, 75.40.Cx, 76.60.-k

I. INTRODUCTION

The field of low-dimensional and geometrically frustrated magnetism is an active area of research in solid state physics. In the last few decades, special emphasis has been laid on low-dimensional spin systems such as chains, square lattices, ladders, especially after the discovery of high-temperature superconductivity in cuprates. In the case of one-dimensional (1D) antiferromagnetic (AF) chains, the scenario becomes even more interesting if in addition to a nearest-neighbor ($nn$) interaction, a frustrating next-nearest-neighbor ($nnn$) interaction is also present. Depending upon the ratio of the $nnn$ to $nn$ coupling ($J_{nnn}/J_{nn}$) in these so-called “Majumdar-Ghosh” (MG) chains (or $J_{nn} - J_{nnn}$ Heisenberg) distinct magnetic phases are formed. For $J_{nnn}/J_{nn} \geq 0.24$, the ground state is spontaneously dimerized with an energy gap in the excitation spectrum.

A number of Cu-based ($3d^9$) AF systems which can be described by the MG (or $J_{nn} - J_{nnn}$ Heisenberg) chain model have been investigated in the past. Some examples are CuCrO$_2$ ($\text{CuCrO}_2$), (N$_2$H$_5$)$_2$CuCl$_3$ and (Cu(ampy)Br)$_2$. Of these, CuCrO$_2$ is thought to be close to the MG point ($J_{nnn}/J_{nnn} \approx 0.5$) and (N$_2$H$_5$)$_2$CuCl$_3$ is close to the quantum critical point ($J_{nnn}/J_{nn} = 0.24$). Surprisingly, it appears that the few vanadium-based ($3d^1$) $S = 1/2$ low-dimensional AF systems that have been investigated have exclusively been either of the dimer type (CuV$_2$O$_4$), and (VO)$_2$P$_2$O$_7$ or ladder type ((VO)$_2$P$_2$O$_7$, CaV$_2$O$_5$). On the other hand, we were unable to find in literature any examples of vanadium-based ($3d^1$) $S = 1/2$ systems described by the $J_{nn} - J_{nnn}$ Heisenberg chain model. We have been exploring low-dimensional oxides with the intention of unraveling novel magnetic properties. It seems interesting to investigate systems where the magnetic exchanges arise from the overlap of the (say) $d_{xy}$ orbitals (via the oxygen $p$) rather than the $d_{x^2-y^2}$ orbitals as in the Cu-based systems. In this paper, we report our studies on the yet unexplored system BaV$_3$O$_8$ via magnetization, heat capacity and $^{51}$V nuclear magnetic resonance (NMR) measurements. The susceptibility $\chi(T)$ data exhibit a broad maximum at 25K signifying short-range order (SRO). The appearance of LRO at $T_N \sim 6$K is evidenced in the susceptibility as well as the heat capacity data. From our $\chi(T)$ data, we infer that the linkages between the magnetic ($S = 1/2$) V$^{4+}$ ions in this compound are like those of an MG chain with $\alpha \approx 2$. The NMR signal from the magnetic $^{51}$V ions could not be observed most probably due to the strong on-site fluctuations of the V$^{4+}$ moment giving rise to a wipeout of the NMR signal. On the other hand, a $^{51}$V NMR signal corresponding to the nonmagnetic V$^{5+}$ nuclei was easily observed. Further, the evolution of its lineshape, spin-lattice relaxation rate $1/T_1$ and spin-spin relaxation rate $1/T_2$ clearly indicates that they are sensitive to the LRO onset and are driven by the fluctuations of the magnetic V$^{4+}$ ions. In the vicinity of LRO, the existence of two components is observed in the spin-spin relaxation data indicating the co-existence of non-LRO and magnetically ordered regions.
II. SAMPLE PREPARATION, CRYSTAL STRUCTURE, AND EXPERIMENTAL DETAILS

BaV₃O₈ is a monoclinic system (space group P2₁/m) containing both magnetic and non-magnetic vanadium ions. Among the three vanadium ions in the unit cell, two are in the V⁵⁺ (S = 0, non-magnetic) state and one is in the V⁴⁺ (S = 1/2) state. The V⁴⁺ ions appear to form chains where the interactions between the magnetic vanadium ions are likely mediated through oxygen (O²⁻) and the non-magnetic V⁵⁺ ions (see Fig. 1). Based on the structure, one expects comparable nn and nnn interactions within the chain. The inter-chain interactions are expected to be weaker. One might, therefore, think of this system as made of coupled MG chains. Since no magnetic data have been reported on BaV₃O₈, we have pursued the problem further and report here the following parameters: (1) the atomic coordinates and occupations are given in Table I, the cell parameters obtained for BaV₃O₈ (Space group: P2₁/m, a = 7.432, b = 5.549, c = 8.200Å, β = 107.207°). The goodness of the Rietveld refinement as defined by the following parameters is R_p = 18.6%, R_w_p = 14%, R_exp = 11.10%, and χ² = 1.58. By simple valence counting, from among the three vanadium ions in the unit cell, two would be in the V⁵⁺ (S = 0, non-magnetic) state and one in the V⁴⁺ (S = 1/2) state. In the unit cell of BaV₃O₈, V⁵⁺ ions form tetrahedra with the oxygen ions whereas the V⁴⁺ ions form pyramids with the oxygen ions.

The various bond angles and bond lengths are given in Table II. The possible interaction paths between the V⁴⁺ ions are illustrated in Fig. 1. The V⁴⁺ ions appear to be arranged in a coupled MG chain-like fashion (along the crystallographic b-direction) and further form corrugated planes as shown in Fig. 1. If we concentrate on an MG chain it can be seen that the V⁴⁺ ions form triangular plaquettes and in a given triangular plaquette the bond distances and the bond angles between any two vanadiums are nearly the same. This suggests a frustrated scenario of an MG chain with comparable J_{nnn} and J_{nn}. The coupling between the V⁴⁺ ions of adjacent chains (J₁) (in a corrugated plane) might be weaker than the intra-chain V⁴⁺-V⁴⁺ interaction since the inter-chain V⁴⁺-V⁴⁺ interaction path consists of one extra oxygen ion. There could also be a weaker V⁴⁺-V⁴⁺ interaction (J₂) in a direction perpendicular to the corrugated plane of the coupled-MG chains. Therefore, based on the structural details alone one might expect SRO in this system.

The temperature dependence of the magnetization M

| Atoms       | Coordinates | Occupancy |
|-------------|-------------|-----------|
|             | x(Å)        | y(Å)      | z(Å)      |
| Ba(2e)      | 0.212       | 0.250     | -0.099    | 1.00   |
| V⁴⁺(2e)     | 0.250       | 0.250     | 0.361     | 1.00   |
| V⁵⁺(2e)     | 0.294       | 0.750     | 0.216     | 1.00   |
| O₁(4a)      | 0.480       | 0.494     | 0.200     | 1.00   |
| O₂(2e)      | 0.432       | 0.750     | 0.476     | 1.00   |
| O₃(4a)      | 0.156       | 0.505     | 0.261     | 1.00   |
| O₄(2e)      | 0.167       | 0.750     | 0.019     | 1.00   |
| O₅(2e)      | -0.171      | 0.250     | 0.319     | 1.00   |
| O₆(2e)      | -0.145      | 0.250     | 0.545     | 1.00   |

Table I: Atomic positions in BaV₃O₈

| Angles       | Description | Value   |
|--------------|-------------|---------|
| V⁴⁺-O²⁻(1)   | intra-chain | 137.07° |
| O²⁻(1)-V⁵⁺(2)| intra-chain | 113.08° |
| V⁵⁺(2)-O²⁻(2)| intra-chain | 147.75° |
| V⁴⁺-O²⁻(3)   | intra-chain | 163.76° |
| O²⁻(3)-V⁵⁺(1)| intra-chain | 101.01° |
| O²⁻(1)-V⁵⁺(2)| intra-chain | 104.89° |
| O²⁻(3)-V⁴⁺(5)| inter-chain | 112.44° |
| V⁵⁺(1)-O²⁻(5)| inter-chain | 83.15°  |
| O²⁻(5)-V⁴⁺(4)| inter-chain | 145.43° |

Table II: Bond angles between various vanadium and oxygen ions in BaV₃O₈

| Bonds       | Description | Value   |
|-------------|-------------|---------|
| V⁴⁺-O²⁻(1)  | intra-chain | 2.01Å   |
| O²⁻(1)-V⁵⁺(2)| intra-chain | 1.71Å   |
| V⁵⁺(2)-O²⁻(2)| intra-chain | 1.74Å   |
| O²⁻(2)-V⁴⁺  | intra-chain | 2.07Å   |
| V⁴⁺-O²⁻(3)  | intra-chain | 1.81Å   |
| O²⁻(3)-V⁵⁺(1)| intra-chain | 1.83Å   |
| V⁵⁺(1)-O²⁻(5)| inter-chain | 1.74Å   |
| O²⁻(5)-O²⁻(4)| inter-chain | 2.78Å   |
| O²⁻(4)-V⁴⁺  | inter-chain | 1.62Å   |

Table III: The bond lengths of the various vanadium-oxygen linkages in BaV₃O₈
Figure 1: Schematic diagram of the coupled-MG chain network formed by the V$^{4+}$ ions. Possible interaction paths between the V$^{4+}$ ions are shown in the figure.

Figure 2: The x-ray diffraction pattern of BaV$_3$O$_8$ is shown along with its Bragg peak positions. The red points are the experimental data, the black curve is the “Fullprof” generated refinement pattern, the green markers are the Bragg peak positions and the blue points represent the difference between the measured and the calculated intensities.

Figure 3: Possible inter-chain interaction paths between the V$^{4+}$ ions

was measured in a magnetic field of 5 kOe in the temperature $T$ range 2 – 300K using a vibrating sample magnetometer (VSM) attached to a Quantum Design Physical Property Measurement System (PPMS). The temperature dependence of the specific heat has been measured in the temperature range of 2 – 270K by using a Quantum Design PPMS. The field-sweep NMR spectra were recorded by means of a home-built phase coherent pulsed spectrometer by sweeping the magnetic field at several fixed frequencies. Typical pulse lengths were 5 and 10 $\mu$s for the $\pi/2$ and $\pi$ pulses, respectively, with a pulse separation of 50 $\mu$s. At each temperature, the area under the spin-echo magnitude was integrated in the time domain and averaged over multiple scans. The $^{51}$V nuclear spin-lattice relaxation was measured by the inversion recovery method using a $\pi$ - $t$ - $\pi/2$ - $\tau$ - $\pi$ pulse sequence with $\tau$ = 50 $\mu$s and variable $t$. To obtain the saturation magnetization $M_0$, the first $\pi$-pulse was switched off every even scan. Subtracting results of odd and even scans one obtains $M_t$ which is generally fit to $M_t = -M_0 + 2M_0 \times e^{-t/T_1}$. In the present case, two components were found in the relaxation behaviour. Consequently, the above equation was modified to accommodate two exponentials with different coefficients. The spin-spin relaxation rate $1/T_2$ measurements were performed in the temperature range of 1.65 – 9.2K at a fixed frequency of 70MHz on the maximum of the spectra (at an applied field 62.6 kOe). The decay of the spin echo (integrated intensity) following a $\pi/2$ - $\pi$ pulse sequence was monitored as a function the pulse separation $\tau$.

III. RESULTS

With decreasing temperature, $\chi$ follows the Curie-Weiss law and shows a broad maximum at about 25K (see Fig.4). With a further decrease in $T$, a sharp drop is observed in $\chi(T)$ at $T_N \approx$ 6K. A Curie-like
upturn is observed at lower temperatures. From the Curie-Weiss fit \( \chi(T) = \chi_0 + C/(T - \theta_{\text{CW}}) \) in the range 80 – 300K, we get the \( T \)-independent susceptibility \( \chi_0 = 5.07 \times 10^{-3} \text{ cm}^3/\text{mol V}^{4+} \), the Curie constant \( C = 0.39 \text{ cm}^3 \text{K}/\text{mol V}^{4+} \), and the Curie-Weiss temperature \( \theta_{\text{CW}} = -26\text{K} \). With \( S = 1/2 \) for the \( V^{4+} \) ion, this Curie constant yields a \( g \)-factor of 2.04 indicating a very small spin-orbit coupling. From \( \chi_0 = 5.07 \times 10^{-3} \text{ cm}^3/\text{mol V}^{4+} \) we obtain the Van Vleck susceptibility \( \chi_{\text{VV}} = \chi_0 - \chi_{\text{core}} = 1.94 \times 10^{-3} \text{ cm}^3/\text{mol V}^{4+} \) where \( \chi_{\text{core}} \) is the core diamagnetic susceptibility equal to \(-1.43 \times 10^{-4} \text{ cm}^3/\text{mol formula unit}\). The broad maximum at 25K might signify the onset of SRO in the MG chains. The second anomaly observed at \( T_N \) (see inset of Fig.4) perhaps evidences the onset of LRO. With the frustration parameter \( f = J_{\text{nn}}/J_s \sim 5 \) it appears that the system is moderately frustrated.\(^{20}\)

The magnetic susceptibility of BaV\(_3\)O\(_8\) was modeled using the exact diagonalization results for the susceptibility \( \chi_{\text{MG}}(g, \alpha, J_{\text{nn}}) \) of a single chain provided by Heidrich-Meissner et al.\(^{21,22}\) (where \( \alpha = J_{\text{nn}}/J_{\text{nn}} \)). Since our experimental results did not match those of the isolated Majumdar-Ghosh chain simulation for different parameters \( \alpha \) and \( J_{\text{nn}} \), we used the expression for a coupled Majumdar-Ghosh chain model, where the inter-chain spin exchange is treated within a mean-field approach\(^{23}\) as eqn.\(^{11}\)

\[
\chi_{\text{coupled MG}}(T) = \left( \chi_{\text{MG}}/(1 - \lambda \chi_{\text{MG}}) \right) + \chi_0 \tag{1}
\]

Here, \( \lambda \) is the mean-field parameter

\[
\lambda = (z_1 J_1 + z_2 J_2)/N_A g^2 \mu_B^2
\tag{2}
\]

where \( J_1 \) and \( J_2 \) are the inter-chain coupling constants within the plane and perpendicular to the plane, respectively, as shown schematically in Fig.4 and Fig.5. The corresponding number of neighbours is \( z_1 \) and \( z_2 \). We found the best agreement of our data with the coupled MG chain model calculations for \( \alpha = 2, J_{\text{nn}}/k_B = 40\text{K} \), and \( \lambda = 21\text{mol/cm}^3\text{cm}^3/\text{mol}\). Taking \( z_1 = z_2 = 2 \) (see Fig.4 and Fig.5) we get \( J_1 + J_2 = 16\text{K} \). We remark in passing that the MG point itself is exemplified by \( \alpha = 0.5 \). The positive sign of the net inter-chain coupling constant indicates a ferromagnetic interaction. The significant inter-chain interactions seem to drive the system to a LRO state at low temperature. Whereas fitting of our bulk susceptibility data are indeed suggestive of a coupled MG chain scenario, we caution that any definitive conclusions need to be backed up by additional work such as neutron scattering measurements, density functional theory calculations to estimate the relative exchange couplings.

The heat capacity data of BaV\(_3\)O\(_8\) are shown in Fig.6 and exhibit a sharp anomaly at about 5.8K. This is close to the transition temperature \( T_N \) measured in \( \chi(T) \). The transition temperature does not shift with \( H \) up to 90 kOe. We have modeled the lattice contribution using the Debye model in the \( T \)-range 60 – 110K. The measured heat capacity could be fit with a combination of two Debye functions of the type given below with coefficients \( C_1 \) and \( C_2 \) (see Fig.6).

\[
C_p = 9 N k_B n (T/\theta_D)^3 \int_0^{\theta_D/T} (x^4 e^x)/(e^x - 1)^2 dx \tag{3}
\]

where \( n \) is the number of atoms in a formula unit, \( N \) is the Avogadro number, \( k_B \) is the Boltzmann constant.
and $\theta_D$ is the relevant Debye temperature. The fit yields Debye temperatures of 180K and 580K and their coefficients 0.25 and 0.52, respectively. Upon subtracting the lattice heat capacity with the above parameters, we obtain the magnetic contribution to the heat capacity $C_m(T)$.

Subsequently, the entropy change ($\Delta S$) was calculated by integrating the $C_m/T$ data (see Fig.6 right inset). The entropy change from about 50K to 2K is about 5.4 J/K which is more than 90% of the value for a $S = 1/2$ system ($Rln(2S + 1)$). The small disparity may be associated with the uncertainty in extracting the lattice contribution. Note also that, upon cooling, most of the entropy decrease has already taken place above $T_N$. This is a fingerprint of strong intra-chain interactions in the system which give rise to SRO. The spectrum is rather broad at $T = 71$ with $T\leq 4K$ which suggests the presence of antiferromagnetic magnons in the ordered state.\footnote{We were unable to detect the NMR signal associated with the magnetic V$^{4+}$-nuclei of BaV$_3$O$_8$. This is because of a strong on-site local moment which naturally couples well with its own nucleus. The fluctuations of this moment are very effective in causing a fast relaxation of the nuclear magnetization. This makes the detection of its NMR signal difficult. In Cs$_2$CuCl$_4$ as well, an NMR signal from the $^{63,65}$Cu nuclei was not detected probably for similar reasons.\footnote{On the other hand, the NMR signal from the nonmagnetic V$^{5+}$ nuclei in BaV$_3$O$_8$ was easily observed. The spectrum is rather broad at 71.5K with the full width at half maximum FWHM of about 0.43 kOe and the total extent of the spectrum of about 1.6 kOe (see Fig.7). The FWHM increases slightly with decreasing temperature down to about 6K and increases drastically below that (see Fig.8) which must be associated with the onset of LRO already indicated in $\chi(T)$ and $C_p(T)$ data. This implies that the V$^{5+}$ nuclei are sensitive to the internal magnetic field arising in this compound. It is shown in Fig.8 that FWHM in LRO phase ($T = 1.85K$) decreases with $H$ although it tends to the value of about 1.1 kOe in zero field which is 2 – 3 times more than the FWHM above $T_N$. Thus the externally applied magnetic field $H$ is not the only source of such a broadening however the $^{51}$V NMR shift does not change much with temperature which indicates that the V$^{5+}$ions are very weakly coupled with the electrons of the magnetic vanadium (V$^{4+}$) ions.

We have measured the recovery of the longitudinal $^{51}$V nuclear magnetization as a function of temperature to probe the low-energy spin excitations. The resulting recovery could be fitted well with a double exponential

$\Delta S = \int_{T_{min}}^{T_{max}} C_m/T dT$

\text{where $T_{min}$ is the minimum temperature for which $C_m$ can be measured, and $T_{max}$ is the maximum temperature.}$

Figure 6: The temperature dependence of specific heat of BaV$_3$O$_8$; the inset on the right displays the magnetic contribution to the heat capacity (black filled circles). The red line represents the lattice heat capacity (see text). The blue line in the lower inset represents the exact diagonalization results of Ref.\footnote{Ref.\footnote{Note that the total heat capacity of the main figure is normalised to the number of atoms (12 per formula unit) whereas the magnetic heat capacity in the inset is normalised to the number of magnetic atoms, i.e., one for each formula unit. The green data points (right axis) show the change of entropy with $T$. The upper inset shows the linear temperature dependence of the magnetic specific heat with $T^3$. The solid line is a linear fit to low temperature data.}}\text{21,22} with ($\alpha = 2, \lambda = 0$). Note that the total heat capacity of the main figure is normalised to the number of atoms (12 per formula unit) whereas the magnetic heat capacity in the inset is normalised to the number of magnetic atoms, i.e., one for each formula unit. The green data points (right axis) show the change of entropy with $T$. The upper inset shows the linear temperature dependence of the magnetic specific heat with $T^3$. The solid line is a linear fit to low temperature data.
(consisting of a short and a long component) at all temperatures. The recovery was found to follow

\[ 1 - M(t)/M_0 = B_Se^{-t/T_1S} + B_Le^{-t/T_1L} + C_1 \]  \hspace{1cm} (4)

Here \( T_{1S} \) and \( T_{1L} \) represent the short and the long components of \( T_1 \), \( B_S \) and \( B_L \) stand for their relative weights respectively and \( C_1 \) is a constant. As seen in the structure, two inequivalent \( V^{5+} \) ions are present in BaV\(_3\)O\(_8\) (Fig. 11). One of them (\( V^{5+}(2) \)) is near the centre of a triangular plaquette and appears coupled to three \( V^{4+} \) ions. The other vanadium (\( V^{5+}(1) \)) seems coupled to two \( V^{4+} \) ions via oxygen. Whereas it is not clear as to the relative magnitudes of the associated transferred hyperfine couplings in the above two cases, the relevant bond angles and bond lengths suggest that the various \( V^{4+}-O-V^{5+} \) couplings may not be too much different from each other. Therefore, the \( V^{5+}(2) \) which is hyperfine coupled to three \( V^{4+} \) might be expected to have a shorter \( T_1 \) compared to that for \( V^{5+}(1) \). The variation of the faster rate \( 1/T_{1S} \) with temperature is shown in Fig. 9 (the slower component has the same qualitative behaviour). As seen from this figure, a distinct anomaly of \( 1/T_1 \) is observed at \( T_N \).

Additionally, we measured the temperature dependence of the transverse decay and obtained the spin-spin relaxation rates \( 1/T_2 \) presented in figure 11. As seen from the raw data at 2.22K in Fig. 11 (right inset), the decay has two components shown by the two dashed lines. Consequently, we have fit the data at each temperature to a double exponential function

\[ M_t = A_se^{-2\pi/T_2S} + A_Le^{-2\pi/T_2L} + C_2 \]  \hspace{1cm} (5)

Here, \( T_{2S} \) and \( T_{2L} \) denote the shorter and the longer components, respectively, \( A_S \) and \( A_L \) stand for their relative weights respectively and \( C_2 \) is a constant. The variation of \( 1/T_{2S} \) and \( 1/T_{2L} \) with temperature is shown in Fig. 10. As seen from Fig. 10, the short component, \( 1/T_{2S} \), exhibits a pronounced \( \sim50\% \) decrease in the vicinity of \( T_N \) while the longer one, \( 1/T_{2L} \), is insensitive to the magnetic ordering. The relative weight of the faster component (\( A_S/A_L \)) decreases monotonically with increasing temperature (as seen in the inset of Fig. 10) varying from about 10 at 1.5K to about 0.1 at about 7.5K. As is evident, at higher temperatures the spin-spin relaxation is dominated by the longer component. There is, therefore, a larger uncertainty in \( T_{2S} \) at higher temperatures. We also note that there is no sharp anomaly in the temperature dependence of \( A_S/A_L \) near \( T_N \). Finally, it seems natural to think that \( A_S/A_L \) represents the relative amount of magnetically ordered regions with respect to non-LRO regions.

Given the above information, it is clear that the \( ^{51}\text{V} \) NMR lineshape in the low-temperature regime is affected by the measurement conditions, in particular, by the pulse separation \( \tau \) between the \( \pi/2 \) and the \( \pi \) pulse. This is clearly seen in Fig. 11 where the \( ^{51}\text{V} \) spectra powder pattern is displayed at fixed frequency and temperature, but for different pulse separations \( \tau \). A broader resonance line is observed for smaller \( \tau \). This might be the reason for the observation of Fig.8 where the FWHM keeps increasing with decreasing temperature without saturation even well below \( T_N \). It is also worth noting that even above \( T_N \), where no ordered moments are expected, a contribution to the width from the shorter \( T_2 \) remains.
We have attempted to obtain the contribution to the lineshape only from the faster $T_{2S}$ component (i.e., the one originated from the magnetic regions) in the following manner. With a large pulse separation like $\tau = 500 \mu s$, the lineshape contains the contribution only from the slow component. This is corrected using $T_{2L}$ and the spectrum from only the long component is obtained at $\tau = 0$. Then with our shortest pulse separation $\tau = 25\mu s$, the spectrum was measured. This contains both the fast and the slow components with dominating fast component at low temperatures. This is corrected for $T_{2S}$ and the spectrum at $\tau = 0$ is obtained. We then subtract the first data set ($T_{2L}$ dominated) from the latter. The resulting spectrum contains the contribution of only the faster $T_2$ component (one can see though that in the temperature region where $A_S/A_L \sim < 1$, this method suffers from a large uncertainty). The resulting spectra at various temperatures are shown in Fig.12. The inset shows the linewidth obtained from such spectra which arises only from the magnetic regions. It is higher than the uncorrected data and also has a sharper variation near $T_N$.

IV. CONCLUSION

In this work we have reported the crystal structure, magnetization, heat capacity, and NMR measurements on a new vanadium-based magnetic system BaV$_3$O$_8$. According to the crystallographic symmetry, the arrangement of the magnetic ions in BaV$_3$O$_8$ is like that of a coupled Majumdar-Ghosh chain with the possibility of three dimensional interactions. We found that the magnetic susceptibility shows a broad maximum at about 25K indicating magnetic short-range order followed by a sharp anomaly at $T_N = 6K$ due to long-range order. The value of the ‘frustration parameter’ ($f = |\theta|/T_N$) ∼ 5 suggests that the system is moderately frustrated. Our susceptibility data above 15K are well described by the coupled Majumdar-Ghosh chain model with the ratio of the $nn$ to $nn$ magnetic coupling $z = 2$ and $J_{nn}/k_B = 40K$. Considering the inter-chain interactions in the
mean-field approach, we obtain the total inter-chain coupling \( J_{\text{int}}/k_B = 16\text{K} \). However, a validation of the above conclusions would need input from techniques such as inelastic neutron scattering, density functional theory calculations, etc. From the magnetic contribution of the heat capacity \( C_m \) we find that upon cooling, most of the entropy decrease has already taken place at temperatures above \( T_N \). This attests to the strong intra-chain interactions in the system which give rise to SRO. Below \( T_N \), both non-magnetic and magnetically ordered regions are found to coexist which, possibly, is characteristic of the inherent frustration in the system. In view of a unique \( V^{4+} \) site in the structure, the coexistence might imply the presence of lattice distortions at low temperatures creating inequivalent magnetic environments. Further work using techniques such as muon spin rotation and neutron diffraction is needed to explore this aspect.

V. ACKNOWLEDGEMENT

Discussions with R.K.Sharma are acknowledged. T.C., A.V.M., A.A.G., and A.V.T. thank the Joint Indo-Russian RFFI-DST Grant 11-02-92707-IND for financial support. W.K. and N.B. kindly acknowledge support from the German Research Society (DFG) via TRR80 (Augsburg, Munich).