Pressure Effect on Magnetization in Quasi-1D Double-Chain material NaV$_2$O$_4$

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Abstract. From the pressure and temperature dependent magnetization measurements, we observed that quasi-1D half-metallic NaV$_2$O$_4$ showed coexistence of field independent antiferromagnetic (AFM) transition at $T_N = 143$ K and two field dependent subphases at $1 \leq H \leq 5$ T. The two characteristic temperatures, $T_{N1}$ and $T_{N2}$ are associated with the two AFM subphases. Under applied field ($H$), $T_N$ and $T_{N1}$ remained almost unchanged, while $T_{N2}$ moved in accordance with the relation $T_{N2} = 117.2 + 8.9H - 3.8H^2$. We have also measured magnetization at $H = 1$ T under different pressure and temperature. $T_N$ and $T_{N1}$ were found to decrease almost linearly with increasing pressure, and the value of $dT_N/dP$ was comparable to that as reported previously. Interestingly, $T_{N2}$ was also found to follow a similar equation under pressure ($P$), namely $T_{N2} = 124.45 + 0.10P - 0.011P^2$. This is considered to be due to the nonlinear variation of the lattice constant along a-axis indicating anomalous spin ordering along this direction (perpendicular to the VO$_6$ double chains) under applied pressure or field.

1. Introduction

NaV$_2$O$_4$ belongs to the CaFe$_2$O$_4$-type orthorhombic structure (space group Pnma) with VO$_6$ double chains formed by a network of edge-sharing VO$_6$ octahedra aligned along the b-axis forming an irregular hexagonal 1-D channel. Magnetic coupling between strong FM intrachain and weaker AF interchain interactions yielded an antiferromagnetic (AFM) transition at $T_N = 140$ K [1]. Moreover, two AFM subphases, existing in the temperature range of 126-130 K at magnetic field $H \geq 1$ T, were gradually suppressed with increasing $H$, and then disappeared at $H > 5$ T [2]. In addition, the powder X-ray diffraction (XRD) measurements of the lattice constant with varying $x$ in the Ca$_{1-x}$Na$_x$V$_2$O$_4$ compound revealed existence of an anomaly along the a-axis. This phenomenon might be regarded as the asymmetric chemical pressure along a-axis caused by Ca$^{2+}$ substitution for Na$^+$ in the Ca$_{1-x}$Na$_x$V$_2$O$_4$ system. It is plausible that pressure and field dependent magnetization studies would reflect this a-axis anomaly associated with the nonlinear spin ordering along this direction. Recent positive muon-spin spectroscopy study also reported two AFM transitions at 140 K and 120 K [3]. These authors also suggested stability of the AF3 subphase under pressure, but field dependent $T_{N2}$ associated with this subphase showed drastic change. So stability on this phase under pressure needs further clarification. In the present work, we carried out careful pressure and temperature dependent magnetization
measurements and tried to clarify the complex nature of magnetic behavior in this interesting quasi 1-D NaV$_2$O$_4$ system.

2. Experiment
The polycrystalline NaV$_2$O$_4$ sample was synthesized by the solid-state-reaction method under high pressure [1]. Hydrostatic pressure dependent magnetization measurement in NaV$_2$O$_4$ were made with increasing pressure up to 14.5 kbar by the piston cylinder self-clamped technique [4]. We used Daphne-7373 oil as a pressure transmitting medium in the cell and the sample was placed in a cell attached with a lead manometer. Applied pressure was determined from the knowledge of the difference of superconducting transition temperatures of lead between ambient and applied pressures. Magnetization measurements were performed at different pressures using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS-XL7).

3. Result and Discussion
Figure 1 shows the temperature dependent magnetization data at $H = 1$ T under different applied pressure. At ambient pressure, an AFM transition is observed at $T_N = 143$ K along with two other small anomalies, the bending point being at $T_{N1} = 135$ K and the kink being at $T_{N2} = 124$ K, respectively. These two anomalies agree with the two antiferromagnetic transitions in the system as reported earlier by Sakurai [2]. The $T_N$ peak moves to the lower temperature region, while the $T_{N1}$ and the $T_{N2}$ anomalies both taper off with increasing $P$. To determine the characteristic temperatures precisely, we plotted derivative of magnetization $d\chi/dT$ as a function of temperature as shown in Fig. 2. Both $T_N$ and $T_{N1}$ are found to decrease almost linearly upon increasing pressure. Interestingly, pressure dependent behavior of $T_{N2}$ is quite different which increases with pressure up to 4 kbar and then showing a broad hump it decreases almost linearly with increasing pressure. On the other hand, the pressure dependent behavior of $T_{N1}$ and $T_{N2}$ can be represented by the linear relations $T_N = 142.9 - 0.52P$ and $T_{N1} = 135.4 - 0.42P$ as shown in Fig. 3. Intriguingly, the decrease of $T_{N2}$ between ambient pressure and up to $P = 14.5$ kbar is around 1 K and follows the nonlinear curve $T_{N2} = 124.45 + 0.10P - 0.011P^2$ (Fig. 3). The height of the kink at $T_{N2}$ shown in Fig. 1 is suppressed gradually with pressure, appears to be due to the suppression of SDW transition under pressure. More interesting to mention that almost similar pressure dependent trend of variation in $T_N$, $T_{N1}$, and $T_{N2}$ is observed with increasing magnetic field, as shown in Fig. 4. which shows that both $T_N$ and $T_{N1}$ vary almost similarly with field while $T_{N2}$ drastically decreases following the nonlinear equation, $T_{N2} = 117.2 + 8.9H - 3.8H^2$ similarly to the case of its variation with pressure as shown in Fig. 3. This indicates that pressure and field have similar effects on the antiferromagnetic transition temperatures and hence have similar effects on the spin structure of this compound. This is also supported from the concentration x dependent variation of the lattice constants (a, b, c) of Ca$_{1-x}$Na$_x$V$_2$O$_4$ where it was found that the nonlinear variation of lattice constant (a) with x [2]. The corresponding variations of (b) and (c) parameters with x are, however, very small.

The above mentioned pressure dependent behavior of $T_N$ also agrees quite well with that observed from $\mu^+\text{SR}$ measurements [3]. However, we also notice from our study that application of pressure (up to 14.5 kbar) cannot stabilize the AF3 phase. This result is in sharp contrast to the suggestion of Sugiyama et al that AF3 phase can be stabilize by applying pressure. This is because, $T_{N2}$ associated with this place is found to change drastically both under applied pressure and magnetic field.
4. Conclusion
The present work shows that both $T_N$ and $T_{N_1}$ decrease with increasing $P$; however, $T_{N_2}$ acts as quadratic function. The AFM transition temperature $T_N$ and the two subphase transitions $T_{N_1}$ and $T_{N_2}$ behave differently under the application of pressure or magnetic field. $T_N$ and $T_{N_1}$ vary almost linearly with $P$ or $H$ while $T_{N_2}$, associated with the subphase AF3, shows drastic variation under the application of $P$ or $H$. This is considered to be due to the nonlinear (anomalous) variation of lattice constant along the (a) direction (perpendicular to the chains). It is thus concluded that the spin ordering along this direction, under pressure or magnetic field, becomes disturbed (disordered asymmetrically) causing abrupt lowering of $T_{N_2}$. Finally, we can conclude that the nonlinear field- and pressure- dependent behavior is due to the anomaly along a-axis spin structure in the so-called 1-D material NaV$_2$O$_4$ and contraction of inter- / intra- chain inhibits the AFM coupling. To realize the origin of magnetism in NaV$_2$O$_4$, further experiments, such as a neutron scattering study under pressure and magnetic field would be interesting.

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![Figure 1](image1.png)  
**Figure 1.** The temperature dependence of dc susceptibility under various pressures at $H = 1$ T.

![Figure 2](image2.png)  
**Figure 2.** Thermal variation of the derivative of magnetization, $d\chi/dT$, at $H = 1$ T under various pressures.

References
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**Figure 3.** The pressure dependent behavior of the transition temperatures $T_N$, $T_{N1}$, and $T_{N2}$. The dashed lines are guided to the eye.

**Figure 4.** The magnetic field dependent behavior of the transition temperatures $T_N$, $T_{N1}$, and $T_{N2}$. The dashed lines are guided to the eye.