Complex magnetic phases in Ca$_{1-x}$Na$_x$V$_2$O$_4$ with 0≤ x ≤ 1

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The crystal structure of Ca$_{1-x}$Na$_x$V$_2$O$_4$ is the same to that of CaFe$_2$O$_4$; that is, V$_2$O$_4$ double-chains, which are formed by a network of edge-sharing VO$_6$ octahedra, align along the b-axis so as to make an irregular hexagonal one-dimensional (1D) channel. Furthermore, since V ions form a zig-zag chain in the V$_2$O$_4$ double-chain, Ca$_{1-x}$Na$_x$V$_2$O$_4$ is expected to exhibit interesting magnetic behavior due to both geometrical frustration and low dimensionality.

Although the ground state of CaV$_2$O$_4$ was thought to be a gapless chiral ordered state, which is predicted for the $S$=1 zig-zag spin system with the competing nearest- and next-nearest-neighbor AF coupling, recent NMR results suggest the existence of a long-range antiferromagnetic (AF) transition with $T_N$=69 K [1]. This is also consistent with past neutron measurements, in which two different AF substructures coexist in the $a \times 2b \times 2c$ AF supercell, and each superstructure is collinear roughly parallel to the b-axis [2].

The other end member, NaV$_2$O$_4$, recently prepared by a high-pressure technique [3], exhibits metallic conductivity down to 2 K, while χ measurements indicates an AF transition with $T_N$=140 K. Magnetic anisotropy measurements using single crystal samples suggest that the interchain interaction is AF, but the intrachain interaction is ferromagnetic (FM). The AF structure of NaV$_2$O$_4$ has, to authors’ knowledge, never been investigated thus far by NMR, neutron, or $\mu^+\text{SR}$ measurements.

We have therefore made a systematic $\mu^+\text{SR}$ experiment on the Ca$_{1-x}$Na$_x$V$_2$O$_4$ system with x=0 - 1 using polycrystalline samples. We found the existence of static magnetic order below 70 K with mainly two different $\mu^+$-spin precession frequency ($\omega_\mu$) signals for CaV$_2$O$_4$, whereas the four $\omega_\mu$’s below 120 K for NaV$_2$O$_4$. Interestingly, the four $\omega_\mu$’s merge into one at $T$ between 120 and 140 K. Combining with the results of the other samples with 0< x <1, we have clarified the complex magnetic phase diagram of Ca$_{1-x}$Na$_x$V$_2$O$_4$.