Complex magnetic phases in $Ca_{1-x}Na_xV_2O_4$ with $0 \leq x \leq 1$

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The crystal structure of $Ca_{1-x}Na_xV_2O_4$ is the same to that of $CaFe_2O_4$; that is, V_2O_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_2O_4 double-chain, $Ca_{1-x}Na_xV_2O_4$ is expected to exhibit interesting magnetic behavior due to both geometrical frustration and low dimensionality.

Although the ground state of CaV_2O_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_2O_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_2O_4 has, to authors' knowledge, never been investigated thus far by NMR, neutron, or μ^+SR measurements.

We have therefore made a systematic μ^+SR experiment on the $Ca_{1-x}Na_xV_2O_4$ system with x=0 - 1 using polycrystalline samples. We found the existence of static magnetic order below 70 K with mainly two different μ^+-spin precession frequency (ω_μ) signals for CaV_2O_4, whereas the four ω_μ's below 120 K for NaV_2O_4. Interestingly, the four ω_μ'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 $Ca_{1-x}Na_xV_2O_4$.

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