Quantum Phase Transition in the Solid Solution

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The structure of (CuCl)LaNb2O7, a metastable phase prepared using an ion-exchange reaction, consists of Cu2+Cl layers sandwiched by non-magnetic double-layered perovskite LaNb5+2O7 blocks. Thus it is regarded as an S = 1/2 quasi-2D system. Previous magnetic studies of this compound revealed the spin-singlet ground state with a zero-field gap of 2.3 meV to the low-lying triplet excited states. The Br-based counterpart (CuBr)LaNb2O7 exhibits collinear anti-ferromagnetic (CAF) order at TN = 32 K characterized by the propagation vector q = (pi, 0, pi). Apparently, the superexchange interactions through Cu-X-Cl (X = Cl, Br) substantially influence their magnetic properties. The solid solution (CuCl1-yBry)LaNb2O7 has been recently prepared to investigate the correlation between the two states.15, 16 However, it should be noted that (CuCl1-yBry)LaNb2O7 is subject to random disorder due to the differing ionic radii of Cl and Br anions and that the CuCl layer inherent to the magnetism is directly disrupted.

Thus we have decided to study the magnetism of (CuCl)LaTa2O7 and the solid solution (CuCl)La(Nb1-xTax)2O7 [1. 2]. A crucial advantage of studying the Nb-Ta system (over the Cl-Br system) is that the CuCl layers are unaffected by the substitution and that Nb and Ta ions have nearly the same ionic radius (0.64 angstrom). We observed persistence of the spin-singlet state in (CuCl)LaNb2O7 up to x ˜ 0.4, accompanied by a slight reduction of the spin gap with increasing x. (CuCl)LaTa2O7 exhibits collinear antiferromagnetic (CAF) order with TN ˜ 7 K, similar to what is observed in (CuBr)LaNb2O7. In the intermediate region (0.4 < x < 1), we observed CAF order with a significantly reduced magnetic moment but with a nearly constant TN, suggesting that the CAF state coexists with the spin-singlet state in agreement with recent MuSR results.

References

[1] A. Kitada et al., submitted to Phys. Rev. B.
[2] Y. J. Uemura et al., submitted to Phys. Rev. B.

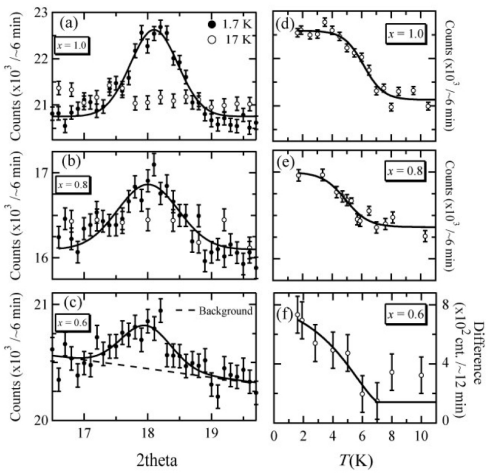

Fig. 1. Neutron diffraction of (CuCl)La(Nb1-xTax)2O7 demonstrating the collinear (pi, 0, pi) magnetic order at low temperatures.