Search for Quantum Criticality in the Spin Tetrahedra System $Cu_2Te_2O_5(Br_xCl_{1-x})_2$

P. Lemmens [a,1], K.-Y. Choi [b], G. Güntherodt [b], M. Johnsson [c], P. Millet [d], F. Mila [e], R. Valenti [f], C. Gros [f], W. Brenig [g]

[a] *Inst. für Metallphysik und Nukl. Festkörperphysik, TU Braunschweig, D-38106 Braunschweig, Germany*
[b] *2. Physikalisches Institut, RWTH Aachen, Templergraben 55, D-56056 Aachen, Germany*
[c] *Department of Inorganic Chemistry, Stockholm Univ., S-10691 Stockholm, Sweden*
[d] *Centre d'Elaboration de Matériaux et d'Etudes Structurales, CEMES/CNRS, F-31062 Toulouse, France*
[e] *Institut de Physique Théorique, Univ. Lausanne, CH-1015 Lausanne, Switzerland*
[f] *Institut für Theoretische Physik, Univ. des Saarlands, D-66041 Saarbrücken, Germany*
[g] *Institut für Theoretische Physik, TU-Braunschweig, D-38106 Braunschweig, Germany*

Abstract

We have investigated the quantum spin system $Cu_2Te_2O_5(Br_xCl_{1-x})_2$ and its tentative proximity to a quantum critical point. This compound allows by changing the stoichiometry x to decrease the unit cell volume, tune the inter-tetrahedra couplings and induce a crossover from an unconventional instability to a Néel state. Substitution studies show a smooth variation of the transition temperature with x. In contrast to the thermodynamic properties the low-energy Raman response drastically changes in a certain concentration range.

Key words: quantum critical point; quantum spin system; Raman scattering

A small variation of certain parameters in a strongly correlated electron system may lead to a drastic change of its ground state and excitation spectrum [1]. This behavior is most prominent in systems with localized spin moments on a two-dimensional (2D) or geometrically restricted antiferromagnetic exchange geometry. The topology of the spin system leads to several nearly degenerate lowest energy quantum mechanical ground states. These states, although having similar ground state energies, are fundamentally different with respect to the evolving spin correlations and excitation spectra. The coupling parameters that promote one of these states instead of others usually lead to a decrease of the corresponding transition temperatures to zero realizing a quantum critical point (QCP). The unconventional scaling induced by the competition of low-energy states is, however, also expected to be observable at finite temperature. Experimental and theoretical investigations of these phenomena have recently gained broader interest and the application of this approach to different model systems led to fundamental insights [1–3]. Quantum criticality may be induced using doping or hydrostatic/chemical pressure on two-leg spin ladder systems with an additional inter-ladder coupling, 2D spin dimers with a frustrating next-nearest neighbor coupling ($SrCu_2(BO_3)_2$) [4] and the square lattice of high temperature superconductors [1].

Recently a compound based on weakly coupled Cu s=1/2 spin tetrahedra has been found that might realize a new class of frustrated quantum spin systems [5–8]. In $Cu_2Te_2O_5(Br_xCl_{1-x})_2$ tetrahedral clusters of Cu^{2+} in a distorted square planar CuO_3X-coordination form a 2D exchange topology in the *ab* plane [5]. Along the *c* axis the tetrahedra align to tubes or chains separated by different Te-O coordinations. With decreasing x the unit cell decreases its volume from 391 to 367 Å^3,

[1] http://www.peter-lemmens.de, E-mail:plemmens@tu-bs.de, Tel.: 0049-(0)531-391-7959, Fax: 0049-(0)531-391-5129

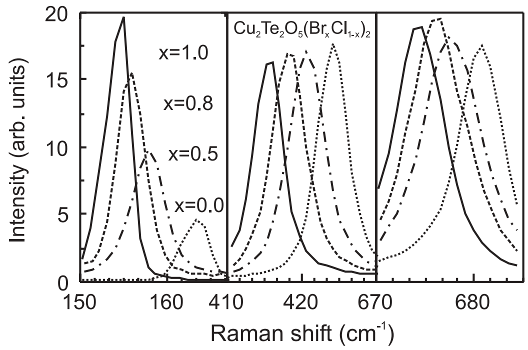

Fig. 1. Systematic shift of selected phonon modes in (cc) polarization with stoichiometry in $Cu_2Te_2O_5(Br_xCl_{1-x})_2$.

corresponding to 7%. In thermodynamic experiments a magnetic instability is observed at T_o=11.4 K that shifts more or less linearly with decreasing x to higher temperatures involving a larger and larger part of the magnetic entropy. Finally, for x=0 T_N=18.2 K is consistent with 3D AF ordering.

In Raman scattering experiments on single crystals a systematic shift of optical phonon modes with composition exists. Selected optical phonon modes are shown in Fig. 1. It is interesting to note that also the intensities vary consistently showing the continuous evolution of physical properties, e.g. the electronic polarizabilities and interband matrix elements involved in the Raman scattering process.

The low-energy excitation spectra of this compound show a more drastic variation with composition. For x=0 a broad and less structured continuum extends from 40 to approximately 100 cm^{-1} without a pronounced temperature dependence [8]. In samples with x=1 a pyramidal-shaped scattering continuum with larger intensity is centered at 61 cm^{-1}. The low-energy regime, however, is dominated by an excitation at 18 cm^{-1} that shows a soft mode behavior consistent with the instability at T_o=11.4 K [6].

For a further progress it is important to understand how the soft mode feature at low energies behaves with decreasing x and associated enhanced couplings of the spin tetrahedra. Therefore a systematic substitution study has been initiated. Respective low energy Raman spectra are shown in Fig. 2. It is obvious from these data that in contrast to the smooth variation of the unit cell volume, phonon frequencies and transition temperatures the evolution of Raman spectra in this system is quite abrupt and detailed. With decreasing x the low energy mode splits into several modes that shift their center of gravity to higher energy. For x=0.7 the low energy excitation spectrum is essentially identical to the previously reported data on x=0 [8].

The shift of the mode to higher energy with decreasing x is consistent with its recent identification [9] as a longitudinal magnon, an oscillation of the local order-parameter close to a QCP. The "critical behavior" found as function of substitution supports this point of view. For such a frustrated spin system [10] the low energy excitation spectrum should change drastically close to a QCP. Although thermodynamic experiments do not directly support this scenario it is suggested that a part of the rich excitation spectrum in this system is based on a remnant of this zero temperature instability.

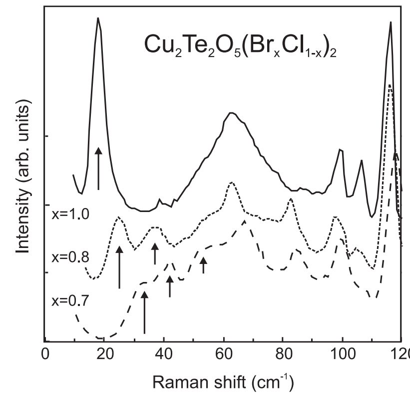

Fig. 2. Low-energy Raman spectra in single crystals of $Cu_2Te_2O_5(Br_xCl_{1-x})_2$ in (cc) polarization. Additional low-energy modes are marked by arrows. The spectra are shifted vertically for clarity.

Acknowledgements

We acknowledge important discussions with H. Kageyama and M. Isobe. This work has been supported by DFG-SP1073.

References

[1] S. Sachdev, Science **288**, (2000) 475.

[2] F. Mila, Eur. J. Phys. **21**, (2000) 499.

[3] P. Lemmens, G. Güntherodt, C. Gros, submitted to Physics Reports (2002).

[4] H. Kageyama, *et al.*, Phys. Rev. Lett. **82**, (1999) 3168.

[5] M. Johnsson, K. W. Törnroos, F. Mila, and P. Millet, Chem. Mater. **12**, (2000) 2853.

[6] P. Lemmens, *et al.*, Phys. Rev. Lett. **87**, (2001) 227201.

[7] W. Brenig and K. W. Becker, Phys. Rev. B **64**, (2001) 214413.

[8] P. Lemmens, *et al.*, Journ. Phys. Chem. Solids **63/6-8**, (2002) 1115.

[9] C. Gros, *et al.*, in preparation (2002).

[10] V.N. Kotov, M.E. Zhitomirsky, O.P. Sushkov, Phys. Rev. B **63**, 064412 (2001).