Unusual criticality of Cu$_2$Te$_2$O$_5$Br$_2$ under pressure

J. Kreitlow and S. Söllow
Institut für Metallphysik und Nukleare Festkörperphysik,
TU Braunschweig, 38106 Braunschweig, Germany

D. Menzel and J. Schoenes
Institut für Halbleiterphysik und Optik, TU Braunschweig, 38106 Braunschweig, Germany

P. Lemmens
Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart, Germany

M. Johnsson
Department of Inorganic Chemistry, Stockholm University, S-10691 Stockholm, Sweden
(Dated: November 6, 2018)

We present measurements of the magnetic susceptibility $\chi(T)$ on Cu$_2$Te$_2$O$_5$Br$_2$ under externally applied pressure. From our data we extract the pressure response of the antiferromagnetic phase transition at $T_0 = 11.6$ K and of the overall magnetic coupling strength. Our experiments indicate that with pressure the overall magnetic coupling strength increases by about 25% with applied pressure of only $\sim 8$ kbar. In contrast, the phase transition temperature $T_0$ is significantly suppressed and not observable anymore at a pressure of already 8.2 kbar.

PACS numbers: 75.10.Jm, 75.40.Cx, 75.40.Gb

The spin-tetrahedra system Cu$_2$Te$_2$O$_5$Br$_2$ belongs to a class of quantum magnets which has been in the focus of intense research efforts recently [2, 3, 4, 5, 6]. Here, the presence of a spin gap through dimerization for a quantum magnet does not lead to a non-magnetic singlet ground state. Instead, based on thermodynamic and spectroscopic techniques an unusual magnetic ground state has been evidenced. Tetragonal Cu$_2$Te$_2$O$_5$Br$_2$ contains clusters of Cu$^{2+}$ with $S = 1/2$ in a distorted square planar CuO$_3$Br coordination (Fig. 1). These tetrahedra form weakly coupled sheets within the crystallographic $a$-$b$-plane. Therefore, this system is ideal to study the interplay between the spin frustration of a tetrahedron with localized low energy excitations and the tendency for a more collective magnetism induced by inter-tetrahedra couplings.

The thermodynamic properties of Cu$_2$Te$_2$O$_5$Br$_2$ are ascribed to two magnetic couplings within the tetrahedra, with the competing exchange constants $J_1$ and $J_2$ [2], and an inter-tetrahedra coupling $J_c$ [9]. As result of the coupling, the system undergoes a phase transition at $T_0 = 11.6$ K. Neutron powder diffraction of Cu$_2$Te$_2$O$_5$Br$_2$ reveals an antiferromagnetically ordered state with a strongly reduced magnetic moment of 0.51(5)$\mu_B$/Cu$^{2+}$ below $T_0$ [5]. On a microscopic level the cause for the phase transition has not unambiguously been resolved [2, 4, 10, 11, 12]. In particular, with the existence of low lying excitations a magnetically ordered state close to quantum criticality has been discussed.

Pressure experiments have proven to be a particularly useful tool to study quantum critical behavior. Therefore, in this work we present a pressure study on Cu$_2$Te$_2$O$_5$Br$_2$. For our experiments we used a CuBe pressure cell in a commercial SQUID magnetometer to measure $\chi(T)$ at pressures up to 8.2 kbar and in external fields up to 5 T for temperatures 2 - 40 K. A powder sample, which has been prepared as described in Ref. [1], was pressed together with GE-Varnish into a pellet,

FIG. 1: A view of the crystal structure of Cu$_2$Te$_2$O$_5$Br$_2$ onto the crystallographic $a$-$b$ (a) and $a$-$c$ plane (b) as illustration for the planar arrangement of the Cu$^{2+}$ tetrahedra.
which was placed in the middle of a teflon tube. The tube was filled with a hydraulic pressure medium (FC-77) and was loaded into the CuBe pressure cell. In Fig. 2 we plot a set of representative measurements on Cu$_2$Te$_2$O$_5$Br$_2$ with our cell for pressures up to 8.2 kbar.

According to ambient pressure experiments [2], the magnetic susceptibility $\chi(T)$ exhibits a broad maximum at about $T_{Max} = 30$ K. This susceptibility maximum represents a measure for the overall magnetic coupling strength, viz., the size of $J_1$ and $J_2$. Below $T_{Max}$ a strong reduction of $\chi$ occurs, as is typical for the onset of antiferromagnetic correlations. The ordering temperature $T_0$ is identified as a step in the temperature derivative $\partial \chi / \partial T$.

Our data at 0.1 kbar closely resemble the ambient pressure behavior from Ref. [2]. Since a step in $\partial \chi / \partial T$ corresponds to a maximum in $\partial^2 \chi / \partial T^2$, we determine $T_0$ at 0.1 kbar from the latter quantity to 11.6 K, in good agreement with Ref. [2] (Fig. 3).

With increasing pressure the phase transition temperature $T_0$ decreases, and we obtain $T_0 = 9.8$ K at 1.5 kbar (Fig. 3). At 3.5 kbar the maximum in $\partial^2 \chi / \partial T^2$ has shifted to 5 K. However, since for both the data taken at 1.5 kbar and 8.2 kbar a similar, but much smaller maximum in $\partial^2 \chi / \partial T^2$ appears, this feature at 5 K might possibly be induced by a changing cooling mode of the SQUID in this temperature range.

Alternatively, it could be argued that for the 3.5 kbar measurement the broad anomaly underlying the peak at 5 K represents a remnant of the antiferromagnetic ordering. In that case the data would indicate a range of $T_0$ between 5 and 8 K. Still, the observation of similar broad anomalies for the higher pressure experiments seems to speak against such interpretation.

In Fig. 3 we summarize the pressure dependence of $T_0$. The error bar at 3.5 kbar reflects the uncertainty about the determination of $T_0$ at this pressure. Altogether, the data suggest a suppression of $T_0$ in the range 5-8 kbar. This statement is supported by the absence of any clear signature of magnetic ordering for the measurement at 8.2 kbar. Hence, our experiments indicate that Cu$_2$Te$_2$O$_5$Br$_2$ is situated in the proximity to a nonmagnetic phase. As the constituting unit is a tetrahedron with antiferromagnetic exchange interaction this phase is suggested to be identical with a short range correlated singlet phase. However also other scenarios have been put forward based on theoretical arguments [11]. Two experimental finding shine further light on the peculiarity of Cu$_2$Te$_2$O$_5$Br$_2$. First is the observation of a related instability with a much higher $T_0=18.2$ K in Cu$_2$Te$_2$O$_5$Cl$_2$ that has a 7% smaller unit cell volume [2]. This compound also has a completely different low energy excitation spectrum in light scattering experiments [3]. The second observation is the evidence for an incommensurate ordering vector of Cu$_2$Te$_2$O$_5$Br$_2$ for $T<T_0$ [3]. Both experimental results imply that the ordering temperature is not only given by a mean field-like inter-tetrahedra coupling and that some additional effect, most probably some antisymmetric Dzyaloshinskii-Moriya (DM) interaction plays some role to establish long range ordering.

Moreover, from $\chi(T)$ we derive the pressure dependence of the susceptibility maximum. It increases from $T_{Max} = 28.5$ K (determined via $\partial \chi / \partial T = 0$ ) at 0.1 kbar applied pressure to $T_{Max} = 40$ K at 8.2 kbar (Fig. 4). This increase indicates a very substantial strengthening of the intra-tetrahedra coupling with applied pressure, yielding an enhancement of 25% at highest applied pressure.

The contrasting pressure response of $T_0$ and $T_{Max}$ is very unusual and likely reflects competing energy scales. If the ordering temperature $T_0$ would be only controlled by the overall magnetic coupling strength, we would ex-
pect an increase of $T_0$ with $T_{\text{Max}}$. Therefore, the decreasing $T_0$ possibly is the result of enhanced frustration $J_1/J_2$ on the tetrahedra. Another scenario would be a weakened inter-tetrahedra coupling $J_c$ with pressure. This however seems unlikely as under pressure the inter-tetrahedra distance decreases which in the absence of structural symmetry modifications should lead to an increase of $J_c$.

In summary, we have performed a pressure study on the susceptibility of Cu$_2$Te$_2$O$_5$Br$_2$. We have determined the pressure response of the antiferromagnetic phase transition temperature $T_0$ and the overall magnetic coupling strength. While we find a strengthening of the magnetic coupling with pressure, attributed to intra-tetrahedra exchange paths, antiferromagnetic order is rapidly suppressed. Tentatively, we relate this behavior to an enhancement of frustration or a weakening of antisymmetric interactions in the system. However, to weight such scenarios Cu$_2$Te$_2$O$_5$Br$_2$ additional thermodynamic and spectroscopic pressure experiments to higher pressure, structural studies under pressure and following theoretical investigations will be necessary. Such work is in preparation.

This work was supported by the Deutsche Forschungsgemeinschaft DFG under projekt number SU/6-1, SPP1073, and INTAS 01-278.

[1] M. Johnsson, K.W. Tornroos, F. Mila, P. Millet, Chem. Mater. 12, 2853 (2000)
[2] P. Lemmens, K.-Y. Choi, E. E. Kaul, C. Geibel, K. Becker, W. Brenig, R. Valentí, C. Gros, M. Johnsson, P. Millet, and F. Mila, Phys. Rev. Lett. 87, 227201 (2001)
[3] P. Lemmens, K.-Y. Choi, G. Güntherodt, M. Johnsson, P. Millet, F. Mila, R. Valentí, C. Gros, W. Brenig, Physica B 329-333, 1049-1050 (2003)
[4] M. Prester, A. Smontara, I. Živković, A. Bilušić, D. Drobac, H. Berger, and F. Bussy, Phys. Rev. B 69, 180401 (2004)
[5] O. Zaharko, A. Daoud-Aladine, S. Streule, A. Furrer, J. Mesot and H. Berger, cond-mat/0405513 (2004)
[6] A.V. Sologubenko, R. DellAmore, H.R. Ott, P. Millet, cond-mat/0406522 (2004)
[7] W. Brenig, Phys. Rev. B 67, 064402 (2003)
[8] R. Valentí, T. Saha-Dasgupta, C. Gros and H. Rosner, Phys. Rev. B 67, 245110 (2003)
[9] C. Gros, P. Lemmens, M. Vojta, R. Valentí, K.-Y. Choi, H. Kageyama, Z. Hiroi, N. V. Mushnikov, T. Goto, M. Johnsson and P. Millet, Phys. Rev. B 67, 174405 (2003)
[10] J. Jensen, P. Lemmens, and C. Gros, Europhys. Letters 64, 689 (2003)
[11] K. Totsuka and H. J. Mikeska, Phys. Rev. B 66, 054435 (2002)
[12] V.N. Kotov, M.E. Zhitomirsky, M. Elhajal, and Frederic Mila, cond-mat/0404674 (2004)