Spin ladder systems have proved to be a rich source of physics over the years. Isolated two-leg \( S = 1/2 \) spin ladders are known to have a spin liquid ground state with \( J \) ladders, \( J \) rungs and \( J \) on the the geometry of inter-ladder coupling but also the inter-ladder coupling is of the order \( J \) 3D coupled ladders \([2, 3]\), a 2D planar array of ladders diagram shown in Fig. 1(a). This is the case for both strong-rung (dimer) limit such as \((\text{C}_{5}\text{H}_{12}\text{N})_{2}\text{CuBr}_{4}\) \([5]\) and \(\text{CaV}_{2}\text{O}_{5}\) \([5, 6]\), which places those systems far from ladder physics. It is not easy to tune \( J_{\text{inter}} \) close to \( J_{c} \), (Dimethylammonium)(3,5-dimethylpyridinium)\( \text{CuBr}_{4} \) was shown to contain isotropic ladders with a strong antiferromagnetic inter-ladder coupling \( J_{\text{inter}}/J \approx 0.32 \), placing it close to the QCP, and displays long-range magnetic ordering at 2 K \([11, 12]\). The observation of a pronounced specific heat anomaly at the magnetic transition indicates that the inter-ladder coupling is too strong to capture a clear signature of the criticality.

Orbital ordering often reduces the magnetic dimensionality of systems \([13]\) due to the anisotropic overlap of orbital wave functions. In \(\text{La}_{2}\text{RuO}_{4} \) the orbital order leads to the formation of spin ladders on a quasi-2D crystal structure \([14]\). Similarly, in \(\text{CuSb}_{2}\text{O}_{6}, \) quasi-1D chains are formed via orbital ordering \([15]\). Since the low dimensionality is provided by orbital ordering while the crystal structure remains quasi-2D or 3D, remnants of 2D and 3D interactions are often not negligibly small and could supply the necessary inter-ladder coupling to approach the QCP.

\(\text{Ba}_{2}\text{CuTeO}_{6} \) with \( S = 1/2 \) \( \text{Cu}^{2+} \) crystallizes in an ordered hexagonal perovskite structure \([16]\). As can be seen in Figures \([1]\) (b) and (d), the crystal structure consists of alternate stacking along the \( c \)-axis of layers with triangular arrangements of \(\text{CuO}_{6} \) octahedra and layers with triangular arrangements of \(\text{TeO}_{6} \) octahedra. The \(\text{CuO}_{6} \) octahedra and the \(\text{TeO}_{6} \) octahedra in neighboring layers are connected alternately by their corners and faces, giving rise to the unit cell containing two \(\text{Cu} \)-layers and two \(\text{Te} \)-layers along the \( c \)-axis. Since \( \text{Cu}^{2+} \) is Jahn-Teller ac-
exchange couplings are indicated by $J_l$ labeled $j$. Inter-ladder couplings are labeled in Ba$_2^+$ arrows. The arrow with label 'J-T' indicates the Jahn-Teller distortion direction. (c) The Cu is highlighted in yellow. The ladders form a stacked layer within the $ab$-plane along the $2$-axis in Ba$_2$CuTeO$_6$. These chains can couple to form ladders via superexchange through oxygen ions. The intra-ladder leg and rung exchange couplings are indicated by $J$ and $J'$ respectively. The diagonal intra-ladder coupling is labeled $j_2$ and the intra-layer inter-ladder couplings are labeled $j_1$ and $j_3$. The inter-layer coupling between ladders through face-sharing TeO$_6$ octahedra is labeled $j_4$. (d) The crystal structure of Ba$_2$CuTeO$_6$ looking onto the $ab$-plane with the same range as in (b). $J$, $J'$, $j_2$ and $j_3$ are indicated by arrows. The top layer of ladders is shown as thick dark blue lines.

In this study we show that the magnetic susceptibility and NMR data on Ba$_2$CuTeO$_6$ are fully consistent with the presence of coupled $S=1/2$ ladders. The sizeable inter-ladder coupling places this compound almost exactly at the QCP. A marginal magnetic transition, lacking clear signatures of a transition in NMR, specific heat and neutron diffraction data, was identified in the magnetic susceptibility $\chi(T)$ which we argue to mirror the presence of strong quantum fluctuations around the QCP.

Polycrystalline samples were prepared from stoichiometric mixtures of dried BaCO$_3$, CuO and TeO$_2$ under flowing oxygen at 950-1000°C. Powder x-ray diffraction and neutron diffraction on HRPD at ISIS confirmed the samples to be of single phase. Single crystals were grown with a flux method based upon that of Köhl et al. using BaCO$_3$, CuO and TeO$_2$. Single crystal x-ray diffraction at $T=296$ K confirmed the structure to be consistent with that reported by Köhl et al. but in a higher symmetry space group of $C2/m$. In $\chi(T)$ for the single crystals and powders no Curie-like contribution could be well fitted. A best estimate of the magnetic impurity content indicates $<0.1\% S=1/2$ impurities, showing that our samples are extremely clean, partly due to full cation ordering caused by the contrasting ionic radii and valences of Cu$^{2+}$ and Te$^{6+}$. Magnetization and specific heat were measured using a Quantum Design MPMS and PPMS respectively. $^{125}$Te NMR ($I=1/2$ and about 7% natural abundance) measurements at a fixed frequency of 55.44 MHz (corresponding to $\mu_0H = 4.12$ T) were performed for $2K \leq T \leq 300$ K using a conventional pulsed NMR technique. LSDA+U calculations were performed for the $C2/m$ structure.
shown in Fig. 2 confirms the hypothesis of Ba$_2$CuTeO$_6$ as a quasi-1D ladder system. The broad overturn around $T \approx 75$ K is characteristic of low-dimensional systems. Fits to the inverse susceptibility for $170 \text{ K}$ to $300 \text{ K}$ gave $\theta_B \approx -113$ K indicating reasonably strong antiferromagnetic interactions and an effective moment of 1.96$\mu_B$/Cu$^{2+}$. $\chi(T)$ is not particularly well fitted by a Heisenberg chain model [24, 25] but it is better described by an isolated two-leg ladder model [5] above $T = 35$ K as can be seen in Fig. 2 (expressions are given in the Supplemental Material (SM) [26]). The ladder model fit, including correction for core diamagnetism $\chi_{dia} = -1.48\times10^{-4}$emu mol$^{-1}$, for $H_{ab}$ gives $J/k_B \approx 86$ K, $J'/J \approx 0.98$, and $g \approx 2.08$, with $\Delta/k_B \approx 40$ K. At low temperatures $\chi(T)$ remains finite and deviates from ladder behavior as will be discussed later.

The $^{125}$Te NMR data supports the spin ladder picture discussed for the $\chi(T)$ data. The $^{125}$Te NMR spectrum of the powder sample is the sum of two components with a 1:1 ratio in agreement with the two inequivalent Te sites present in the crystal structure (see Fig. 1). Each spectrum could be consistently fitted using an anisotropic Knight shift tensor. The color contour plot of the $^{125}$Te NMR intensity in the temperature-magnetic field plane, shown in Figure 3(a), represents the temperature dependence of the Knight shift, which agrees well with $\chi(T)$. The temperature dependence of the spin-lattice relaxation rate (1/$\tau$) for arrays of single crystals can be fitted reasonably well with an Arrhenius type of behavior ($1/T_1 = A\exp(-E_a/k_B T)$) at high temperature above 14 K, as shown in Figure 3(b), giving an activation energy $E_a/k_B \approx 50$ K, close to the expected spin gap $\Delta$ in the 'pure' spin ladder limit extracted from $\chi(T)$.

Estimates of effective exchange coupling constants from LSDA+U band structure calculations support the previously described ladder picture. The total energy $E(\mathbf{q}, \phi)$ was calculated as a function of a wave vector $\mathbf{q}$ and of an angle $\phi$ between spins of two Cu$^{2+}$ ions in the $T = 300$ K monoclinic $C2/m$ unit cell for a number of spin-spiral structures. Effective exchange coupling constants were evaluated by fitting $E(\mathbf{q}, \phi)$ to a classical Heisenberg model. We obtain antiferromagnetic leg coupling $J = 33.8 \text{ meV}$ and rung coupling $J' = 33.0 \text{ meV}$ for $U = 5 \text{ eV}$, yielding almost isotropic $J'/J \approx 0.98$ in excellent agreement with the $\chi(T)$ fit. There is roughly a factor of four difference between $J$ obtained experimentally and that from the $U = 5 \text{ eV}$ calculation. The calculated $J$ parameters are found to scale down with increasing $U$ approximately as $1/U$ and the agreement with the experimental values are not unreasonable given the uncertainties present. As expected, the other parameters are smaller than $J$ and $J'$. The diagonal coupling $j_1$ is antiferromagnetic with $j_1/J \approx 0.05$. There are three main inter-ladder couplings (see Fig. 1(b-d)). $j_2$ and $j_3$ are between ladders in the same plane (parallel to the $ab$-plane) whereas $j_4$ couples ladders between planes through

![Fig. 2](image-url)  

**FIG. 2.** (Color online) $\chi(T)$ for Ba$_2$CuTeO$_6$ single crystal arrays with the magnetic field of $\mu_0 H = 1 \text{ T}$ applied parallel (upper panel) and perpendicular (lower panel) to the $ab$-plane. The dashed and solid lines indicate chain and ladder model fits respectively. The inset to the upper panel shows the low temperature region for both orientations along with the high field behavior for $H_{ab}$. The inset to the lower panel shows $M(H)$ of Ba$_2$CuTeO$_6$ for $H_{ab}$.

![Fig. 3](image-url)  

**FIG. 3.** (Color online) (a) Contour plot of the spin echo intensity of the $^{125}$Te-field-sweep NMR spectra as a function of temperature (NMR frequency: 55.44 MHz) for a polycrystalline sample. 

The dotted line is a guide to the eye. (b) Semi-log plot of $1/T_1$ versus $1/T$. Solid lines correspond to the behavior $1/T_1 = A\exp(-E_a/k_B T)$ at high temperatures.
FIG. 4. (Color online) (a) The specific heat $C$ of single crystal Ba$_2$CuTeO$_6$ plotted as $C/T$ vs. $T^2$. The dashed line indicates an approximate lattice $T^2$ term as described in the text. (b) The estimated magnetic specific heat $C_{mag}(T)$ for Ba$_2$CuTeO$_6$ plotted as $C_{mag}/T$ vs. $T$. The inset shows the estimated magnetic entropy $S_{mag}(T)$ as a percentage of $R \ln 2$ up to $T = 20 K$.

The described lack of specific heat anomaly at the marginal phase transition implies that the peak is smeared out by quantum critical fluctuations and indeed we detect indications of unquenched magnetic entropy at low temperatures. As seen in the inset to Fig. 4(a), $C/T$ vs. $T^2$ is not linear, even below 5 K. A best estimate of the phononic $T^3$ lattice contribution is indicated as a dashed line in Fig. 4(a). For the non-magnetic analogue Ba$_2$ZnTeO$_6$, $C/T$ vs. $T^2$ is completely linear below 5 K [29] and the Debye temperature of Ba$_2$CuTeO$_6$ ($\theta_D \approx 323$ K), estimated from the dashed line in Fig. 4, agrees reasonably well with that of Ba$_2$ZnTeO$_6$ ($\theta_D \approx 377$ K). These facts indicate that the dashed line in Fig. 4(a) is a reasonable estimate of the lattice contribution and that there is an additional contribution to the specific heat other than the lattice which can naturally be ascribed to $S=1/2$ spins. The magnetic specific heat, $C_{mag}(T)$ was estimated by subtracting the aforementioned magnetic $T^3$ term from the total specific heat, the result is shown in Fig. 4(b). $C_{mag}(T)$ was then integrated to provide an estimate of the magnetic entropy $S_{mag}(T)$, shown in the inset to Fig. 4(b), which is approximately 4.5% of $R \ln 2$ at 17 K. Despite the presence of such a large magnetic entropy at low temperature, no clear anomaly is present in $C_{mag}(T)$ at $T_{mag} \approx 16$ K and the entropy is quenched only gradually to $T = 0$ K. We argue that the large magnetic specific heat observed around $T_{mag} \approx 16$ K represents the quantum fluctuations associated with the close proximity to the QCP.

In conclusion, we have discovered that Ba$_2$CuTeO$_6$ is a clean, site ordered system with ‘hidden’ antiferromagnetic $S =1/2$ spin ladders due to ferro-orbital ordering on the Cu$^{2+}$ site. The very weak signature of order in $\chi(T)$, without any corresponding anomaly in specific heat, neutron diffraction or NMR points to the conclusion that Ba$_2$CuTeO$_6$ is almost precisely at the QCP for the antiferromagnetic $S =1/2$ weakly coupled ladder system, where quantum fluctuations dominate. Compared to other systems close to the QCP, such as (Dimethylammonium)(3,5-dimethylpyridinium)CuBr$_4$, in which clear thermodynamic signatures of order are seen, Ba$_2$CuTeO$_6$ may provide a better opportunity to study the quantum criticality of the weakly coupled ladder system.
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