Experimental report

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Title: Magnetic order in highly frustrated KCeS2

Research area: Physics

This proposal is a new proposal

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Samples: KCeS2

Instrument	Requested days	Allocated days	From	To
D1B	2	2	25/09/2020	28/09/2020

Abstract:

Here we propose to study the magnetic structure in KCeS2. The system belongs to the delafossite family, offering disorder free, perfect triangular layers, which in combination with spin-1/2 moments is a model system for studying quantum spin liquid (QSL). Complete characterization confirmed the excellent quality of the sample, although revealed a magnetic order at temperatures below 400 mK. With the frustration factor of the order of 200, this system appears on the edge between conventional order and quantum disorder. Careful examination of the low-energy physics will shed light on the nature of QSL and on subtleties of interaction it depends on.

Experiment Title

Magnetic order in highly frustrated $KCeS_2$ (#5-31-2704, 24–27 September, 2020)

Proposer

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Report

Introduction

The family of the insulating delafossite compounds with the general formula ABX_2, where A is a monovalent alkali metal, B is a trivalent rare-earth element, and X is a bivalent anion, attracted considerable interest of the condensed-matter community after a series of recent publications [1–4]. It has shown that two members of the family, $NaYbS_2$ and $NaYbO_2$, manifest properties of a quantum spin liquid (QSL) state, such as the absence of the long-range magnetic order down to 50 mK, together with the presence of significant correlations between localized spins. The delafossite insulators fulfill all requirements essential for QSL. Structurally, they consist of alternating triangular layers of cations A and B separated by anions X. The triangular lattice offers highly frustrated foothold for localized AFM-coupled spins hosted by the B-site. The separating layers confine the magnetism to a 2D triangular-lattice plane. Both factors promote quantum fluctuations, which destroys the classical order and advances QSL state.

Experimental configuration and results

Our delafossite powder consisted of small crystals with sizes up to 0.5 mm. This sample was placed in a Cu can to ensure good thermal conductivity and cooled down using a dilution refrigerator. It was measured with a rotation around the vertical axis within 180° with a 1° step on the D1B (CRG) high-intensity two-axis powder diffractometer at ILL, France. Neutrons

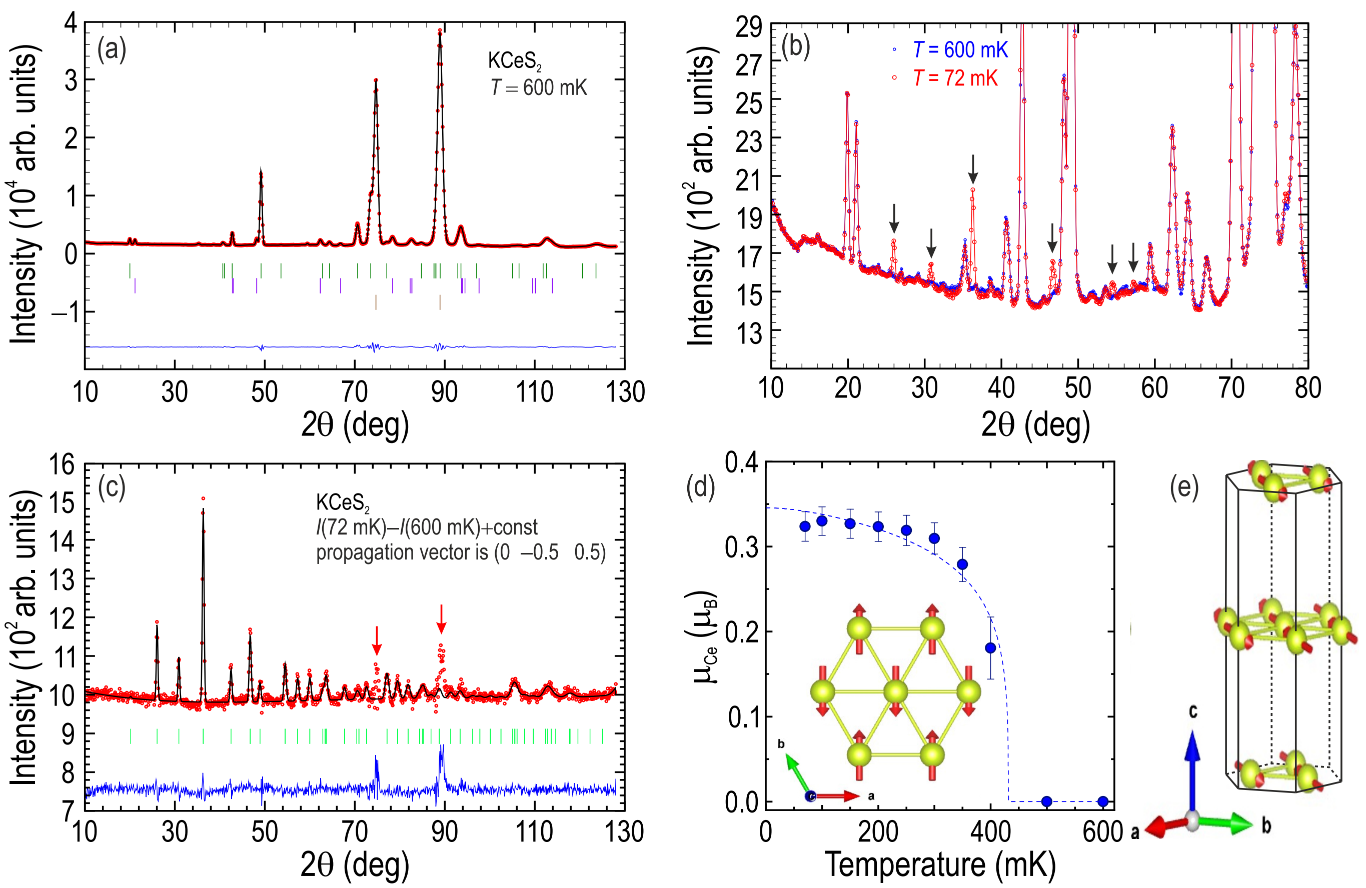

Fig. 1: Neutron powder diffraction data. (a) Scattered neutron intensity at $T = 600$ mK as a function of 2θ fitted to the rhombohedral $R\bar{3}m$ space group. The fit includes CeO_2S as an impurity phase and elemental Cu from the sample environment. Green, purple and brown marks denote peaks from $KCeS_2$, CeO_2S and Cu, respectively. (b) Scattered neutron intensity as a function of 2θ. Blue and red marks are data measured at 600 and 72 mK, respectively. Black arrows show the magnetic peaks, which appear below T_N. (c) The difference of intensities measured at low and high temperatures. Red arrows show the imperfect subtraction of strong structural Bragg reflections. (d) Temperature dependence of the ordered magnetic moment. The refined magnetic structure in the ab plane is shown in the inset. (e) The resulting magnetic structure of $KCeS_2$.

with the wavelength $\lambda = 2.52$ Å were selected using a pyrolytic graphite (002) monochromator. We were interested in the low temperature measurements below 1 K, because $T_N \approx 400$ mK, and good temperature stability.

Neutron powder diffraction data at $T = 600$ mK as a function of 2θ are shown in Fig. 1(a). Crystal structure of antlerite from neutron powder diffraction data was fitted by the rhombohedral $R\bar{3}m$ space group. Obtained low-temperature cell parameters from Rietveld refinement are $a = 4.222(3)$ Å and $c = 21.837(9)$ Å. Green, purple and brown marks denote peaks from the main $KCeS_2$ phase, CeO_2S impurity phase, and Cu can, respectively. The refinement suggests that the CeO_2S impurity phase constitutes about 15% of the sample. This phase is expected as a result of decomposition of $KCeS_2$ in the presence of oxygen. The appearance of magnetic peaks below 400 mK, shown in Fig. 1(b) by black arrows, correlates with the anomaly observed in the magnetic specific heat measurements [5] at the same temperature, corresponding to long-range antiferromagnetic ordering. Rietveld refinement of the magnetic signal in Fig. 1(c) reveals an antiferromagnetic structure with the commensurate propagation vector $(0\ -\frac{1}{2}\ \frac{1}{2})$. The spins lie in the ab plane, oriented perpendicularly to the nearest-neighbor Ce-Ce bonds, as shown in the inset to Fig. 1(d) and in Fig. 1(e). This is consistent with the theoretically proposed "stripe-yz" order [6, 7]. The ordered magnetic moment on Ce^{3+} is about $0.32(1)\mu_B$.

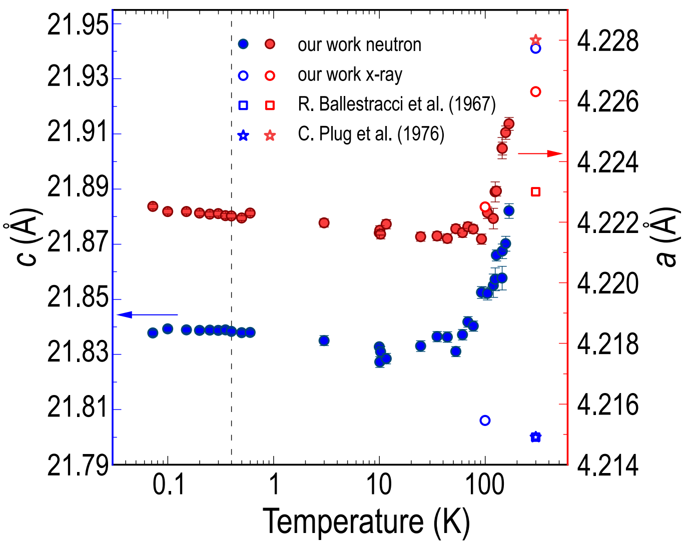

Fig. 2: Temperature dependence of the a (red) and c (blue) lattice parameters. Dashed line shows the antiferromagnetic ordering temperature.

Temperature dependence of the ordered magnetic moment in Fig. 1(d) follows order-parameter behavior with the transition temperature of approximately 435 mK, which is slightly higher than what was previously observed in thermodynamic measurements [5]. The ordered moment saturates below 300 mK, which may be a consequence of poor temperature stabilization in the powder sample close to the base temperature of the dilution refrigerator.

From the refinement of structural lattice parameters, we also obtained the temperature dependence of lattice constants, which is plotted in Fig. 2. Both lattice parameters show conventional (positive) thermal expansion above 100 K, whereas below this temperature a crossover to weakly negative thermal expansion is observed. The switching temperature of the thermal expansion behaviour coincides with the absolute value of the Curie-Weiss temperature and is likely due to magnetostrictive effects that become effective far above the Néel temperature due to the strong frustration in the system. Moreover, we observe no magnetostriction anomaly at the magnetic ordering temperature (dashed vertical line). Absolute values of the cell parameters have only minor deviations a from previously published values.

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