Experimental report

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Council: 4/2021

Title: Low-energy spin waves in the triangular-lattice antiferromagnet KCeS$_2$

Research area: Physics

This proposal is a new proposal

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Samples: KCeS2

Instrument	Requested days	Allocated days	From	To
IN5	3	3	31/08/2021	03/09/2021

Abstract:

Rare-earth delafossites were recently proposed as promising candidates for the realization of an effective S = 1/2 quantum spin liquid on the triangular lattice. In contrast to the most studied triangular-lattice antiferromagnet YbMgGaO4, which is known for very strong structural disorder due to site intermixing, Yb- and Ce-based delafossites NaYbS2, NaYbO2, and KCeS2 realize structurally perfect triangular layers with practically no distortions. In NaYbS2 and NaYbO2, magnetic order is absent down to mK temperatures, suggesting that these compounds may realize the long-sought spin-liquid ground state that can be destroyed by magnetic field that stabilizes long-range order. In contrast, KCeS2 was recently shown to develop magnetic order below 0.4 K already in zero field. We have recently established the magnetic structure of this ordered phase from neutron diffraction and are now proposing to measure spin-wave excitations to estimate the magnon band width and the essential magnetic interactions in the effective magnetic Hamiltonian of the system. Our goal is to understand the origin of the ordered state and why it is present in KCeS2 but not in the isostructural Yb compounds.

Experiment Title

Low-energy spin waves in the triangular-lattice antiferromagnet $KCeS_2$ (# 4-01-1718, 31.08-03.09.2021)

Proposer

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Report

Introduction

Rare-earth delafossites were recently proposed as promising candidates for the realization of an effective $S = \frac{1}{2}$ quantum spin liquid (QSL) on the triangular lattice [1–5]. In contrast to the most actively studied triangular-lattice antiferromagnet $YbMgGaO_4$ [6–11], which is known for very strong structural disorder due to site intermixing [12–14], $NaYbS_2$, $NaYbO_2$, and $KCeS_2$ delafossites realize structurally perfect triangular layers with practically no distortions. In $NaYbS_2$ and $NaYbO_2$, magnetic order is absent down to mK temperatures, suggesting that these compounds may realize the long-sought QSL ground state, which can be destroyed by a magnetic field that stabilizes long-range order. Their magnetic excitation spectrum features a continuum of fractionalized excitations [5, 15] similar to the one observed earlier in $YbMgGaO_4$ [11]—a fingerprint of an emerging QSL ground state. In contrast, $KCeS_2$ was recently shown to develop magnetic order below 400 mK in zero field [16]. This offers a possibility to investigate the variations in magnetic Hamiltonians among the delafossites that are responsible for the stability of the spin-liquid state.

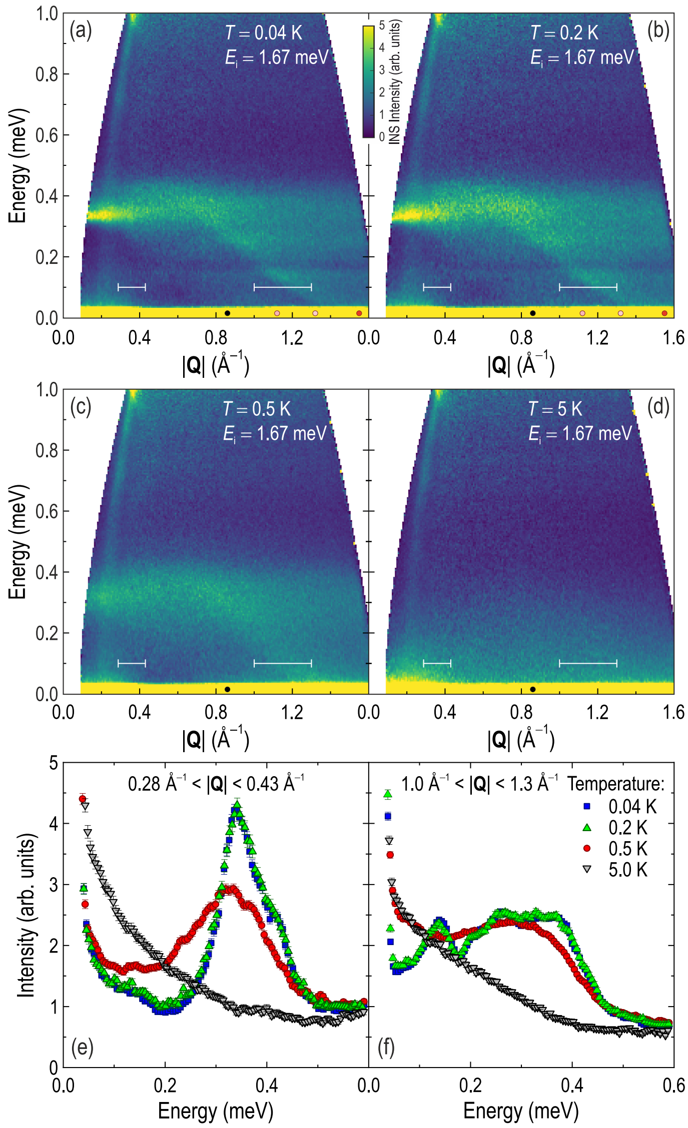

Fig. 1: (a)—(d) Time-of-flight maps with $E_i = 1.67$ meV at different temperatures, where back and red dots show the structural and magnetic peaks, respectively, with the yellow region representing the oversaturated elastic peak. White lines show integration ranges in **Q** for energy cuts shown in (e) and (f) panels, respectively.

Experimental configuration and results

The goal of our proposed experiment was measurements of low-energy magnetic excitations in the ordered state of $KCeS_2$, which we expected to represent spin waves emanating from the magnetic Bragg peaks. Because $KCeS_2$ single crystals cannot be grown in sufficient sizes for neutron spectroscopy, we performed cold-neutron TOF measurements on a powder sample. Because of the expected low energy scale of the signal, the measurement was only feasible on a cold-neutron spectrometer such as IN5.

The phase-pure powder sample of $KCeS_2$ with a mass of 4.16 g was synthesized by our collaborators. We characterised it with a combination of x-ray and neutron diffraction, magnetic, thermodynamic, μSR, and neutron CEF measurements. Crystal structure of this sample is described by the rhombohedral $R\bar{3}m$ space group with $a = 4.222$ Å and $c = 21.837$ Å cell parameters. The magnetic structure below $T_N = 0.4$ K at zero magnetic field is "stripe-yz" order with a commensurate propagation vector $(0\ -\frac{1}{2}\ \frac{1}{2})$ and the ordered magnetic moment on Ce^{3+} of $0.32(1)\mu_B$.

The delafossite powder was measured with E_i = 1.67 meV on the disk chopper time-of-flight (TOF) spectrometer IN5 at ILL, France. The sample was placed in a thin-walled Cu container and cooled down to 40 mK using a dilution refrigerator. We were interested in measuring low-energy excitations at base temperature and above $T_N = 0.4$ K.

Magnetic Bragg peaks appear below the transition temperature in Fig. 1(a,b) at the same positions that were observed earlier in our neutron powder diffraction data [17]. The low-energy spin excitations are seen as a sharp, nearly nondispersive band at 0.34 meV with an intensity maximum at $\mathbf{Q} = 0$. This behavior is not typical for conventional spin waves, where the intensity is expected to be maximized at the magnetic ordering vector. Fig. 1 (e) and (f) show the temperature evolution of spectra with integrated $\mathbf{Q}$ between 0.28 to 0.43 and 1.0 to 1.3 Å^{-1}, respectively. On closer examination, in Fig. 1 (e,f) we see two peaks in the low-$\mathbf{Q}$ region below 1 Å. At higher $|\mathbf{Q}|$, a third peak appears around $E = 0.14$ meV as a result of a spin gap seen as a dip in the $\mathbf{Q}$-integrated spectrum, which is seen in Fig. 1(a,b) as a horizontal dark line. The broad magnetic signal in the inelastic channel is still visible at a temperature of 0.5 K that is slightly above the magnetic transition [Fig. 1(c)] but it disappears at 5 K [Fig. 1(d)]. The temperature evolution of the inelastic features can also be observed in Figs. 1(e,f).

In summary, neutron time-of-flight spectroscopy of low-energy excitations in the ordered state of $KCeS_2$ reveals an unusual spin-wave band with an intensity maximum at $\mathbf{Q} = 0$, unlike in the isostructural $KCeO_2$ [18] and $KYbSe_2$ [19] that show more typical spin-wave bands emanating from the antiferromagnetic ordering vector. A collaboration with theorists is under way to understand this unusual behavior, and data on single crystals would be desirable to get a better understanding of the origin of three features in the INS spectrum and to determine their dispersions.

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