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Anomalous elastic behavior associated with magnetic ordering in Cu_2OSeO_3 observed using resonant ultrasound spectroscopy

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1. Introduction

Cu_2OSeO_3 has attracted considerable interest since the discovery of the skyrmion phase in the magnetic phase diagram [1)]. In addition to the skyrmion phase, Cu_2OSeO_3 exhibits multiple magnetic phases in response to temperature and magnetic-field variations below the transition temperature of $T_c \sim 60$ K [2)]. Typically, Cu_2OSeO_3 experiences a series of transitions as the magnetic field increases, progressing from a multi-domain helical structure through a single-domain conical structure, culminating in a field-polarized collinear ferrimagnetic structure. A fluctuation disordered phase has been observed in a temperature interval of ~1 K immediately below the transition point [2)]. The skyrmion phase emerges in a narrow temperature and magnetic field range near T_c. In contrast to other skyrmion host compounds, Cu_2OSeO_3 is an insulating material that exhibits magnetoelectric coupling, leading to the distinctive properties of skyrmions [3,4)].

Another important aspect of Cu_2OSeO_3 is the strain coupling associated with magnetic ordering. The development of ferrimagnetic order requires a symmetry reduction from a paramagnetic cubic structure to a rhombohedral structure because ferrimagnetism is incompatible with cubic magnetic symmetry [5)]. However, no traces of structural distortion from cubic lattice geometry have been found down to 10 K or in magnetic fields up to 14 T [6)]. In addition, there are no reports of ferroelastic twinning and ferroelectric behavior that would be indicative of a change to rhombohedral lattice geometry, implying that Cu_2OSeO_3 undergoes only magnetic instability. These findings render Cu_2OSeO_3 an important model system for understanding the pure coupling between strain and magnetic ordering, particularly skyrmion.

The influences of strain on the magnetic properties of Cu_2OSeO_3 was investigated with the primary objective of controlling the helimagnetic and skyrmion phases by applying external pressure. T_c increases approximately linearly with hydrostatic pressure at a rate of ~0.3 K/kbar [7)], whereas it barely changes under uniaxial stress up to 101 MPa [8)]. Uniaxial stress or directional pressure components of an applied quasihydrostatic pressure can significantly enhance the stability of the helical and skyrmion phases. These observations of the close relationship between stress and magnetic phase stability provide evidence for magnetoelastic coupling in Cu_2OSeO_3.

Evans et al. [9)] confirmed that although any magnetoelastic coupling in Cu_2OSeO_3 is weak, it indeed gives rise to small but measurable changes in elastic properties at some of the known magnetic transitions. They investigated the overall elastic and anelastic behavior using resonant ultrasound spectroscopy (RUS), focusing on a temperature–magnetic field region well below T_c. In contrast, the primary objective of the present study was to investigate strain coupling with the skyrmion phase by collecting detailed RUS data in a dense region of magnetic phases near T_c. The present RUS results provide not only complementary information for the results of Evans et al. [9)] but also insights into the elastic and anelastic behavior associated with magnetic ordering in Cu_2OSeO_3.

2. Methods

The same Cu_2OSeO_3 single crystal as used in the study of Evans et al. [9)] was used in this study. It has an irregular polygon shape with a pair of growth faces parallel to {111} and mass 1.2 mg. RUS data were collected in low-temperature and magnetic field environments using facilities in Cambridge, whose details have been described elsewhere [10)]. The sample was held lightly between piezoelectric transducers such that the applied magnetic field was parallel to [111]. Resonance spectra were systematically collected in automated sequences of varying temperature in a constant magnetic field or varying field at constant temperature, where the variable temperature and field ranged from 30 to 80 K and 0 to 60 mT, respectively. Elasticity change was examined from the evolution of the square of the resonance frequency (f^2) rather than the elastic constants due to the irregular shape of the Cu_2OSeO_3 crystal. As in conventional RUS, the anelastic property was evaluated in terms of the inverse mechanical quality factor (Q^{-1}).

3. Results and Discussion

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The RUS data obtained in this study newly revealed [11]:

- small anomalies in the temperature dependence of f^2 and Q^{-1} near 59 K in zero field and in a 2 mT field
- hysteretic variations in the elastic property between heating and cooling over the temperature interval 41–58 K
- elastic softening associated with the helical–conical transition
- small discontinuities in the evolution of f^2 between 50 and 58 K
- difference in elasticity between the helical and skyrmion phases upon heatings
- typical frequency dependence of the peak temperature of the Debye-like loss peak centered at ~ 40 K
- Debye-like loss peaks near 50 and 60 K
- strain effects demarcating the stability field boundaries of the fluctuation disordered phase
- small anomalies associated with the helical–conical transition in the evolution of f^2 with magnetic field
- the dependence of elastic anomalies associated with the helical–conical transition on the way in which experiments are conducted

One interesting finding is that the skyrmions in Cu_2OSeO_3 are weakly pinned, if at all, by strain effects. This suggests that the skyrmions show glassy behavior, where the equilibrium state is never reached. In fact, the trend of reducing f^2 values during the full sequence of increasing and decreasing the field at 56 and 56.5 K resembled glassy behavior seen in other systems [12], where there appears to be a continuous drift in the values with time rather than any systematic dependence on magnetic field (**Fig. 1**). The hypothesis that the skyrmions experience very low viscosity, i.e. with liquidlike behavior, appears to match up with their large size compared with twin walls and their extremely weak intrinsic magnetoelastic coupling. Research questions that could be asked include whether weak magnetoelastic coupling is a fundamental requirement for the formation of a skyrmion lattice.

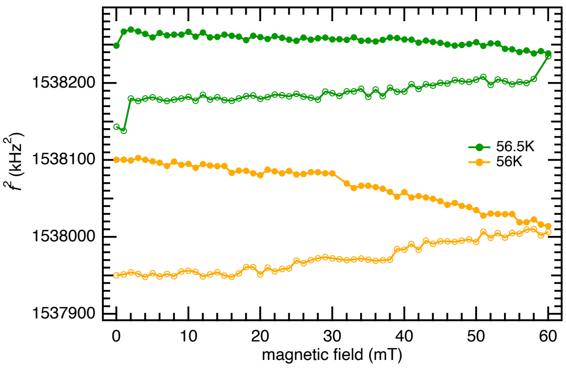

Fig. 1 Evolution of f^2 values for resonances near 1240 kHz as a function of increasing (filled symbols) and decreasing (open symbols) magnetic field at 56 and 56.5 K. f^2 values have been multiplied by arbitrary scaling factors.

4. Conclusions

The most obvious finding of this study is that any anomalies in the elastic and anelastic properties associated with magnetic ordering in Cu_2OSeO_3 are small. The new RUS data suggested that elastic and anelastic anomalies can demarcate the stability field boundaries of the skyrmion phase and fluctuation disordered phases, which are generally quite diffuse. In addition, the measurements of elastic properties with variable fields characterized the glassy behavior of skyrmions in terms of strain coupling.

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