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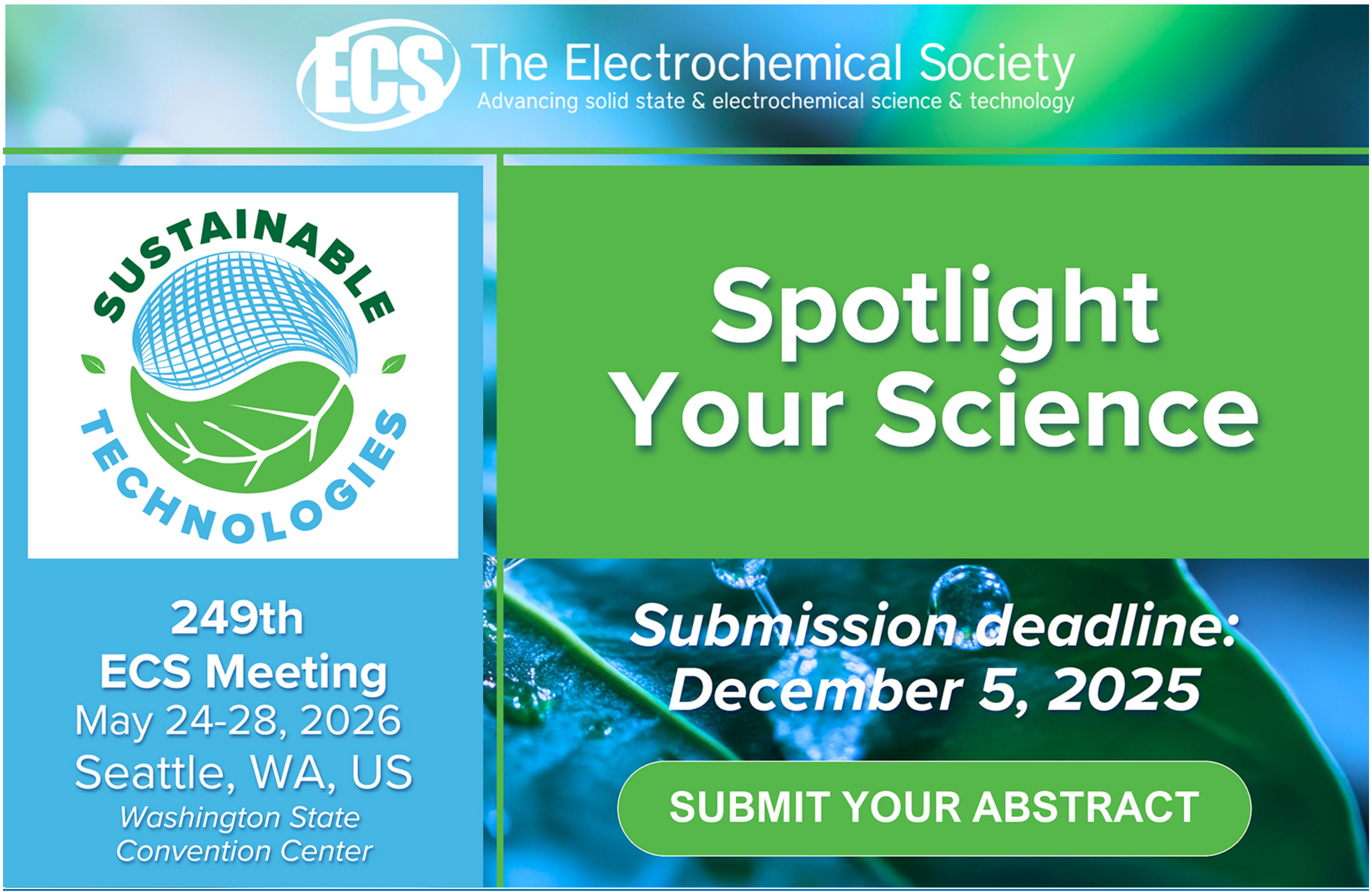

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Ultrasound velocity measurements in orbital-degenerate frustrated spinel MgV_2O_4

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Abstract. Ultrasound velocity measurements of the orbital-degenerate frustrated spinel MgV_2O_4 are performed in the disorder-free high-purity single crystal which exhibits successive structural and antiferromagnetic phase transitions, and in the disorder-introduced single crystal which exhibits spin-glass-like behavior. The measurements reveal coexisting two types of anomalous temperature dependence of the elastic moduli in the cubic paramagnetic phase: Curie-type softening with decreasing temperature, and softening with a characteristic minimum with decreasing temperature. These elastic anomalies should respectively originate from the coexisting orbital fluctuations and spin-cluster excitations.

1. Introduction

Vanadate spinel AV_2O_4 (A = Zn [1], Mg [2] and Cd [3]) is a geometrically frustrated magnet which undergoes a cubic-to-tetragonal structural transition at a temperature T_s and an antiferromagnetic (AF) transition at a low temperature T_N. For AV_2O_4, it is considered that the lowering of the lattice symmetry by the structural transition at T_s leads to the release of the frustration by the AF transition at T_N lower than T_s. Thus the interplay of spin, orbital, and lattice degrees of freedom should play a crucial role for the release of frustration in AV_2O_4. We are interested in the frustrated phase (the cubic paramagnetic (PM) phase) of AV_2O_4. And we performed ultrasound velocity measurements in the magnesium vanadate spinel MgV_2O_4. This compound exhibits successive phase transitions of a cubic-to-tetragonal structural transition at $T_s = 65$ K and an AF transition at $T_N = 42$ K [2]. For MgV_2O_4, it is known that a small amount of disorder suppresses the structural and magnetic phase transitions, and induces spin-glass-like behaviour at low temperatures [4]. In the present study, we performed the measurements in the disorder-free high-purity single crystal with the successive phase transitions at T_s = 65 K and T_N = 42 K, and in the disorder-introduced single crystal which exhibits the spin-glass-like behaviour below $T_f = 12.5$ K.

2. Experimental

The ultrasound velocity measurements were performed in two different types of MgV_2O_4 single crystals grown by the floating-zone method: the disorder-free high-purity single crystal with the successive structural and AF transitions at $T_s = 65$ K and $T_N = 42$ K, named here as “ordered MgV_2O_4”, and the disorder-introduced single crystal with the spin-glass-like behavior below T_f= 12.5 K in which ~3% of V atoms in the octahedral sites are substituted by Mg atoms, named here as "disordered MgV_2O_4" [4]. Temperature (T) dependence of the ultrasound velocity was measured at T from 10 K to 150 K with magnetic field H||[110] up to 7 T in all the symmetrically independent elastic moduli in the cubic crystal: compression modulus C_{11}, tetragonal shear modulus (C_{11}-C_{12})/2, and trigonal shear modulus C_{44}. The configuration of propagation ***k*** and polarization ***u*** of the sound wave for each elastic mode is summarized in Table 1.

Table 1. Elastic modulus of cubic crystal and the corresponding configuration of sound wave with propagation *k* and polarization *u*.

Elastic modulus	Sound wave	Propagation ***k***	Polarization ***u***
Compression modulus C_{11}	Longitudinal wave	[001]	[001]
Tetragonal shear modulus (C_{11}-C_{12})/2	Transverse wave	[1-10]	[110]
Trigonal shear modulus C_{44}	Transverse wave	[001]	[110]

3. Results and discussion

Figures 1(a), (b) and (c) respectively show T dependence of the compression modulus C_{11}, the tetragonal shear modulus (C_{11}-C_{12})/2, and the trigonal shear modulus C_{44} in the ordered MgV_2O_4 with H = 0. All the elastic modes exhibit a jump at T_s and a discontinuous change at T_N, as marked by arrows in Figs. 1(a), (b), and (c). In the cubic PM phase ($T > T_s$), C_{11} and (C_{11}-C_{12})/2 exhibit huge Curie-type (~ -1/T) softening with decreasing T, which should be a precursor to the cubic-to-tetragonal lattice distortion at T_s. As shown in the insets to Figs. 1(a) and (b), this Curie-type softening is independent of H || [110] , and thus should be driven by the coupling of lattice to orbital fluctuations which is hardly affected by the spin sector [5, 6, 7, 8]. On the other hand, C_{44} in the cubic PM phase ($T > T_s$) exhibits non-monotonic softening with decreasing T: softening with convex curvature in ~80 K < T < ~150 K and Curie-type softening in T_s < T < ~80 K. As shown in the inset to Fig. 1(c), the softening in C_{44} is sensitive to H || [110]: the non-monotonic softening with concave curvature in the H

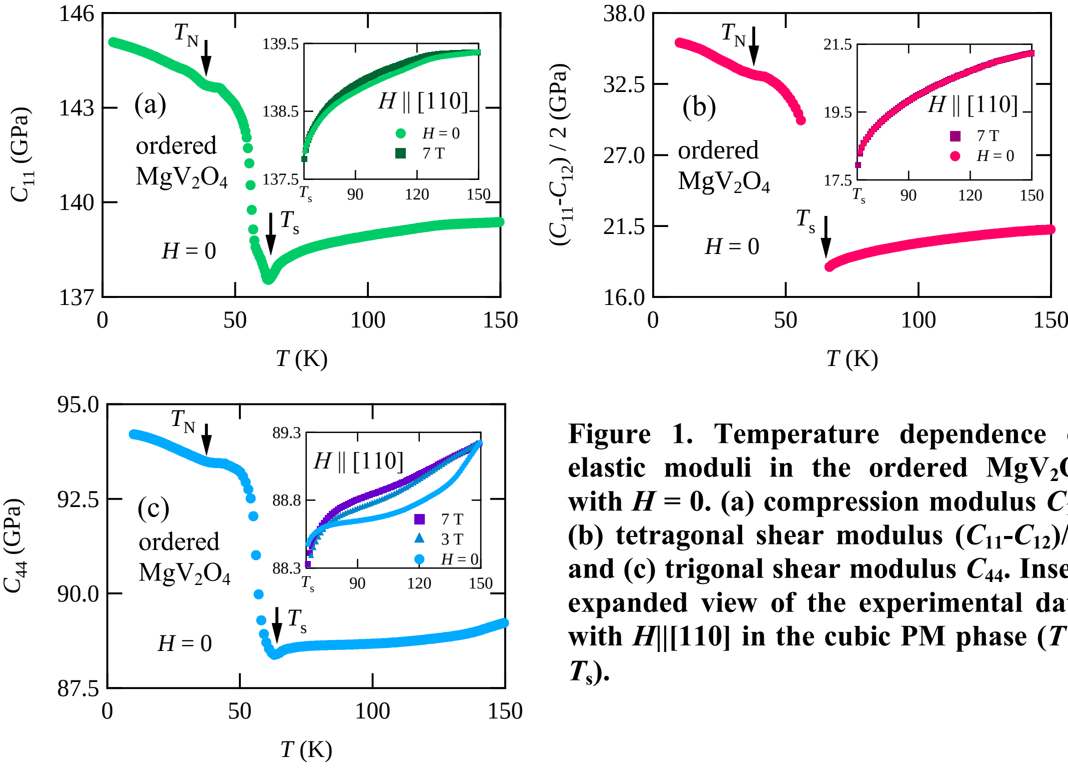

Figure 1. Temperature dependence of elastic moduli in the ordered MgV_2O_4 with H = 0. (a) compression modulus C_{11}, (b) tetragonal shear modulus (C_{11}-C_{12})/2, and (c) trigonal shear modulus C_{44}. Inset: expanded view of the experimental data with H||[110] in the cubic PM phase (T > T_s).

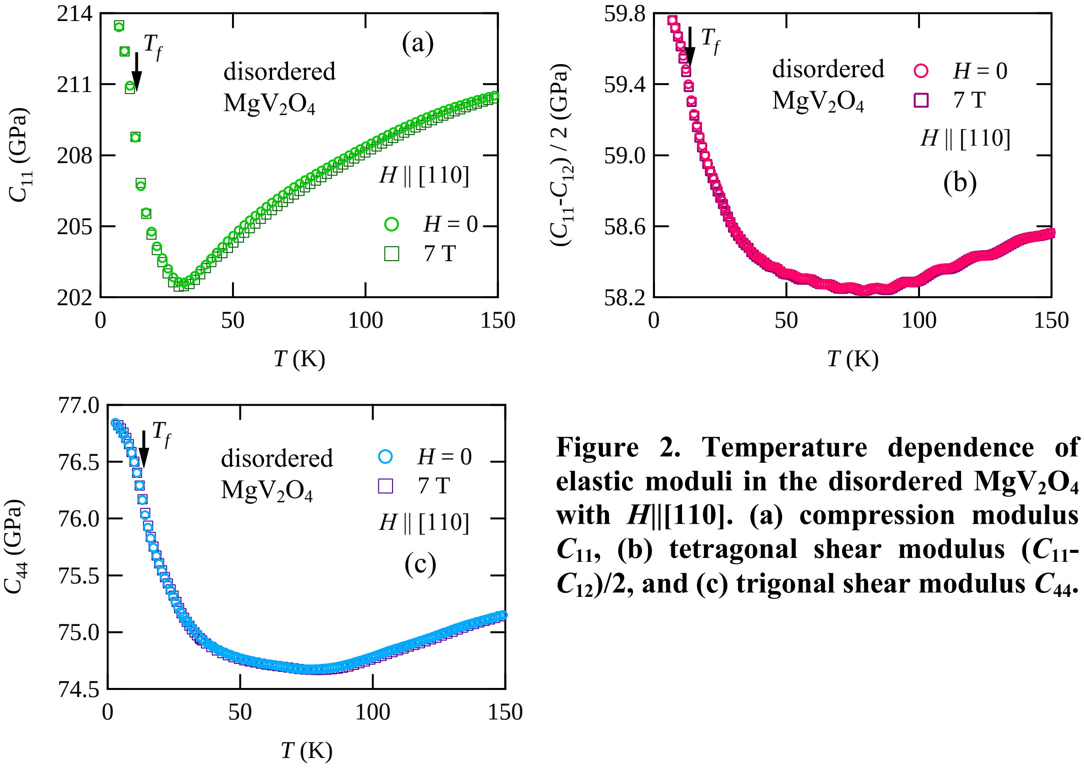

Figure 2. Temperature dependence of elastic moduli in the disordered MgV_2O_4 with H||[110]. (a) compression modulus C_{11}, (b) tetragonal shear modulus $(C_{11}$-$C_{12})/2$, and (c) trigonal shear modulus C_{44}.

= 0 data becomes closer to the Curie-type softening in the 7 T data. Thus, taking into account the H insensitivity of the Curie-type softening in C_{11} and $(C_{11}$-$C_{12})/2$, the non-monotonic softening in C_{44} should observe a superposition of H-sensitive concave T dependence and H-insensitive Curie-type softening. Furthermore, subtracting the component of the Curie-type softening from the observed non-monotonic softening in C_{44}, another component observed as the concave T dependence should be characterized as a softening with minimum with decreasing T. Such a softening-with-minimum anomaly is usually observed as a result of the coupling of lattice to magnetic excitations [9, 10, 11]. Taking into account that inelastic neutrons scattering experiments in AV_2O_4 observed spin-cluster excitations in the cubic PM phase [1], the softening with minimum in C_{44} of the ordered MgV_2O_4 should originate from the coupling of lattice to the spin-cluster excitations.

Figures 2(a), (b) and (c) respectively show T dependence of C_{11}, $(C_{11}$-$C_{12})/2$, and C_{44} in the disordered MgV_2O_4 with H || [110] which behaves differently from that in the ordered MgV_2O_4 shown in Figs. 1(a), (b), and (c). At T_f marked by arrows in Figs. 2(a), (b), and (c), all the elastic modes exhibit a small increase in slope. In the cubic PM phase ($T > T_f$), more notably, all the elastic modes in the disordered MgV_2O_4 exhibit the H-insensitive softening with minimum with decreasing T which should be driven by the coupling of lattice to the spin-cluster excitations.

We here note that the disordered MgV_2O_4 exhibits the absence of the Curie-type softening, namely the absence of a precursor to structural transition, which is compatible with the absence of structural transition in the disordered MgV_2O_4. Taking into account the presence of the Curie-type softening in the ordered MgV_2O_4 as shown in Fig. 1, the present study reveals that not only the structural transition but also its precursor (the Curie-type softening) in MgV_2O_4 is sensitively suppressed by disorder. On the other hand, the softening with minimum is observed in both the ordered and the disordered MgV_2O_4 indicating that the spin-cluster excitations are robust against disorder. Therefore the results in the present study strongly suggest the coexistence of the disorder-sensitive orbital fluctuations and the disorder-robust spin-cluster excitations in MgV_2O_4.

We note here that the component of the softening with minimum in C_{44} of the ordered MgV_2O_4 seen in the inset to Fig. 1(c) is sensitive to H, whereas that in C_{11}, $(C_{11}$-$C_{12})/2$, and C_{44} of the disordered MgV_2O_4 respectively seen in Figs. 2(a), (b), and (c) is insensitive to H. This difference in the H-sensitivity should also arise due to disorder, where the response of the spin-cluster-lattice coupling to H is quenched by the introduction of disorder. The detailed mechanism for this disorder effect remains to be elucidated. For instance, the excitation in MgV_2O_4 might correctly be the orbital-spin-cluster excitation, and its change to the orbital-cluster excitation might occur by the introduction of disorder.

4. Summary

We performed ultrasound velocity measurements of MgV_2O_4 in the disorder-free high-purity single crystal which exhibits successive structural and AF phase transitions, and in the disorder-introduced single crystal which exhibits spin-glass-like behaviour. The measurements reveal coexisting two types of anomalous T dependence of the elastic moduli in the cubic PM phase: Curie-type softening with decreasing T, and softening with a characteristic minimum with decreasing T. These elastic anomalies should respectively originate from the coexisting disorder-sensitive orbital fluctuations and disorder-robust spin-cluster excitations.

5. Acknowledgement

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