Acoustic evidence for dynamical molecular-spin state without undergoing magnetic phase transition in spin-frustrated ZnFe$_2$O$_4$

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(Dated: April 16, 2015)

Ultrasound velocity measurements were performed on a single crystal of spin-frustrated ferrite spinel ZnFe$_2$O$_4$. In this cubic crystal, all the symmetrically-independent elastic moduli exhibited softening with a characteristic minimum with decreasing temperature. This elastic anomaly suggests a coupling between lattice deformations and molecular-spin excitations. The present study also revealed that the elastic anomalies, normally driven by the spin Jahn-Teller phase transition and its precursor, are absent in ZnFe$_2$O$_4$, suggesting that the spin-lattice coupling cannot play a role in relieving frustration within this compound. The magnetic properties of ZnFe$_2$O$_4$ are expected to be fully governed by the dynamical molecular-spin state.

PACS numbers: 72.55.+s, 75.20.-g, 75.40.Gb, 75.50.Xx

I. INTRODUCTION

Cubic spinels $AB_2O_4$ with magnetic $B$ ions have attracted considerable interest in light of the geometrical frustration which is inherent in the $B$-site sublattice of corner-sharing tetrahedra (pyrochlore lattice). One of the most extensively studied spinel systems is chromite spinels $ACr_2O_4$ with $A = Mg$ and Zn, for which the magnetic properties are fully dominated by the Jahn-Teller (JT)-inactive Cr$^{3+}$ with spin $S = 3/2$ (Fig. 1(a)) residing on the pyrochlore network.$^2$ $ACr_2O_4$ with Weiss temperature $\Theta_W \simeq -390$ K undergoes an antiferromagnetic (AF) ordering at $T_N \simeq 13$ K along with a cubic-to-tetragonal structural distortion.$^2$ For AFe$_2$O$_4$ with $A = Zn$ and Cd are another JT-inactive spinel system with Fe$^{3+}$s showing a high spin of $S = 5/2$ (Fig. 1(b)). For AFe$_2$O$_4$ with $\Theta_W \simeq 120$ K ($A = Zn$) and $-50$ K ($A = Cd$), neutron scattering experiments in the high-purity single crystals observed neither long-range magnetic ordering nor a structural transition down to low temperature (1.5 K) although an AF-transition-like anomaly occurs in the magnetic susceptibility at $T^* \simeq 13$ K (Fig. 1(c)). Thus, the frustrated magnetism of AFe$_2$O$_4$ should be different in nature from that of ACr$_2$O$_4$.

For ACr$_2$O$_4$, the magnetostructural phase transition is explained by the spin-JT mechanism via spin-lattice coupling, where local distortions of the tetrahedra release the frustration in the nearest-neighbor AF interactions.$^4,10$ In the frustrated paramagnetic (PM) phase of ACr$_2$O$_4$, inelastic neutron scattering (INS) experiments provided evidence of quasielastic magnetic scattering, indicating the presence of strong spin fluctuations because of spin frustration.$^3,11-13$ Whereas spin-JT fluctuations are expected to exist in the PM phase of ACr$_2$O$_4$, this quasielastic mode involved the fluctuations of AF hexagonal spin molecules (AF hexamers) in the pyrochlore lattice (Fig. 1(d)).$^{14,15}$ Recent ultrasound velocity measurements of MgCr$_2$O$_4$ suggested the coexistence of spin-JT fluctuations and gapped molecular-spin excitations in the PM phase, which is compatible with the recent observation of finite-energy molecular-spin excitations in high-resolution INS experiments in the PM phase of this compound.$^{16}$

For AFe$_2$O$_4$, whereas spin-lattice ordering is absent down to low temperature, magnetic diffuse scattering, arising possibly from the dynamical molecular-spin state, was observed in the INS experiments.$^{2,8}$ Thus, in the absence of the spin-JT effect, the frustrated magnetism in AFe$_2$O$_4$ is expected to be governed by the dynamical molecular-spin state. For ZnFe$_2$O$_4$, the observed diffuse scattering was attributed to fluctuations of the AF twelve-membered spin molecules (AF dodecamers illustrated in Fig. 1(e)).$^{14}$ The formation of the different types of spin molecules in between ZnFe$_2$O$_4$ (the AF dodecamers) and ACr$_2$O$_4$ (the AF hexamers) is considered to arise from the difference in the dominant exchange paths, specifically, the third-neighbor AF interactions $J_3$ with additional nearest-neighbor ferromagnetic $J_1$ for ZnFe$_2$O$_4$ but AF $J_1$ for ACr$_2$O$_4$ respectively. For CdFe$_2$O$_4$, the INS experiments produced a scattering pattern, which resembles that of ACr$_2$O$_4$, indicative of the dominant AF $J_1$.

Notably, molecular-spin excitations were observed in the INS experiments in the frustrated magnets of not only ACr$_2$O$_4$ and AFe$_2$O$_4$ but also HgCr$_2$O$_4$, GeCo$_2$O$_4$, LiV$_2$O$_4$, and Tb$_2$Ti$_2$O$_7$, where the types of spin molecules vary from compound to compound depending on the dominant exchange path. These observations imply that the dynamical molecular-spin state can universally emerge in the frustrated magnets.

In this paper, we present an analysis of ultrasound velocity measurements for the zinc ferrite spinel ZnFe$_2$O$_4$. The sound velocity or the elastic modulus is a useful probe enabling symmetry-resolved thermodynamic information to be extracted from frustrated magnets.$^{14,21,25}$
As mentioned earlier, the observed elastic anomalies in magnesium chromite spinel MgCr$_2$O$_4$ strongly suggested the coexistence of spin-JT fluctuations and molecular-spin excitations in the PM phase. The present study finds elastic anomalies in ZnFe$_2$O$_4$ that provide strong evidence for the emergence of a dynamical molecular-spin state without undergoing a magnetic phase transition.

II. EXPERIMENTAL

The ultrasound velocity measurements were performed on a high-purity single crystal of ZnFe$_2$O$_4$ grown by the flux method. Figure 1(c) shows a plot of the magnetic susceptibility of the ZnFe$_2$O$_4$ single crystal as a function of temperature, where the AF-transition-like anomaly occurs at $T^* \sim 13$ K. The ultrasound velocities were measured using the phase-comparison technique with longitudinal and transverse sound waves at a frequency of 30 MHz. The ultrasonic waves were generated and detected by LiNbO$_3$ transducers glued to the parallel mirror surfaces of the crystal. The measurements were performed at temperatures from 2 K to 300 K for all the symmetrically-independent elastic moduli in the cubic crystal, specifically, compression modulus $C_{11}$, tetragonal shear modulus $C_{44}$, and trigonal shear modulus $C_{44}$. The respective measurements of $C_{11}$, $C_t$, and $C_{44}$ were performed using longitudinal sound waves with propagation $k||[001]$ and polarization $u||[001]$, transverse sound waves with $k||[110]$ and $u||[110]$, and transverse sound waves with $k||[110]$ and $u||[001]$. The sound velocities of ZnFe$_2$O$_4$ measured at room temperature (300 K) are 6480 m/s for $C_{11}$, 2930 m/s for $C_t$, and 3740 m/s for $C_{44}$.

III. RESULTS AND DISCUSSION

Figure 2(a)-(c) presents the temperature dependence of the elastic moduli, $C_{11}(T)$, $C_t(T)$, and $C_{44}(T)$ in ZnFe$_2$O$_4$. The insets depict the compressive, tetragonal, and trigonal lattice deformations of the cubic crystal associated with the propagation $k$ and polarization $u$ of the sound waves. On cooling from room temperature (300 K) to $\sim 100$ K, all the elastic moduli exhibit ordinary hardening consistent with the background $C_{11}^0(T)$ taken from an empirical evaluation (dotted curves in Fig. 2(a)-(c)). However, below $\sim 100$ K, all the elastic moduli exhibit an anomalous temperature variation.

The elastic anomalies in the JT-inactive magnets like ZnFe$_2$O$_4$ are attributed to magnetoelastic coupling acting on the exchange interactions. In this mechanism, the exchange striction arises from a modulation of the exchange interactions by ultrasonic waves:

$$H_{exs} = \sum_{ij}[J(\delta + u_i - u_j) - J(\delta)]S_i \cdot S_j. \quad (1)$$

Here $\delta = R_i - R_j$ is the distance between two magnetic ions, and $u_i$ is the displacement vector for the ion $R_i$. If a sound wave with polarization $u$ and propagation $k$ has form $u = u_0 \exp[i(k \cdot r - \omega t)]$, where $u_0$ and $\omega$ are its amplitude and frequency, respectively, the exchange striction of Eq. (1) is rewritten as:

$$H_{exs} = \sum_i \left( \frac{\partial J}{\partial \delta} - u_0 \right) k \cdot \delta (S_i \cdot S_{i+4}) e^{i(k \cdot R_i - \omega t)}. \quad (2)$$

Here the exponential is expanded to first order because $k\delta \ll 1$. Eq. (2) implies that both the longitudinal and transverse sound waves can couple to the spin system via the exchange striction mechanism, which depends on the directions of both polarization $u$ and propagation $k$ relative to the exchange path $\delta$. 
Similar to ZnFe$_2$O$_4$, the softening-with-minimum elastic anomaly in $C_T(T)$ is also observed in other frustrated magnets of AC$_2$O$_4$ (e.g., SrCu$_2$(BO$_3$)$_2$, GeCo$_2$O$_4$ and MgV$_2$O$_4$) for MgCr$_2$O$_4$ in the PM phase ($T > T_N$) exhibits not only softening-with-minimum in $C_{44}(T)$ from molecular-spin excitations but also a huge Curie-type $-1/T$ softening in $C_{11}(T)$ and $C_4(T)$ which is a precursor to the spin-JT transition. This coexistence of two types of elastic anomalies in MgCr$_2$O$_4$ infers the coexistence of molecular-spin excitations and spin-JT fluctuations. For ZnFe$_2$O$_4$, in contrast, $C_T(T)$ exhibits solely softening-with-minimum, as is clearly seen in the expanded view of $C_T(T)$ (Fig. 3(a)-(c)) [open circles, from Fig. 2(a)-(c)], which infers the presence of molecular-spin excitations but the absence of spin-JT fluctuations.

We also note here that, whereas $C_T(T)$ for MgCr$_2$O$_4$ exhibits a discontinuity at $T_N$ (Fig. 2(d)-(f)), $C_T(T)$ for ZnFe$_2$O$_4$ exhibits no discontinuity at $T^*$ (Fig. 3(a)-(c)). Thus, the experimental $C_T(T)$ for ZnFe$_2$O$_4$ suggests that the AF-transition-like anomaly at $T^*$ in the magnetic susceptibility seen in Fig. 1(c) is not a phase transition. This inference is compatible with the absence of long-range magnetic ordering revealed by the neutron scattering experiments. Regarding the elastic properties of ZnFe$_2$O$_4$, the continuity in elasticity at $T^*$ (the absence of a phase transition) is compatible with the absence of Curie-type softening (the absence of a precursor for the magnetostructural transition), indicating that the spin-lattice coupling in ZnFe$_2$O$_4$ is not strong enough for the spin-JT effect to occur. The precise nature of the magnetic state of ZnFe$_2$O$_4$ below $T^*$ remains to be discovered. Freezing of the spin molecules is a possibility that might occur at $T^*$ in ZnFe$_2$O$_4$.

As is clear from a comparison between $C_T(T)$ for ZnFe$_2$O$_4$ (Fig. 2(a)-(c)) and $C_{44}(T)$ for MgCr$_2$O$_4$ (Fig. 2(f)), the softening in $C_T(T)$ begins to occur below $\sim 50$ K or $\sim 80$ K in ZnFe$_2$O$_4$ but above 300 K in MgCr$_2$O$_4$. As discussed later in conjunction with Eq. (3) and Table I, the reason for the softening occurring at lower temperatures for ZnFe$_2$O$_4$ relative to MgCr$_2$O$_4$ is because the coupling is weaker between the lattice deformations and molecular-spin excitations. Additionally, as is also clear from a comparison between $C_T(T)$ of ZnFe$_2$O$_4$ (Fig. 3(a)-(c)) and $C_{44}(T)$ of MgCr$_2$O$_4$ (Fig. 2(f)), the former exhibits its minimum at $\sim 4$ K or $\sim 6$ K, which is lower than the minimum point of $\sim 50$ K in the latter. As also discussed later in conjunction with Eq. (3) and Table I, the lower temperature at which the minimum point occurs for ZnFe$_2$O$_4$ relative to that for MgCr$_2$O$_4$ is due to the smaller gap associated with its molecular-spin excitations.

Softening-with-minimum in $C_T(T)$ driven by the molecular-spin excitations is generally explained as the presence of a finite gap for the excitations, which is sensitive to strain. Assuming a singlet-triplet excitation gap $\Delta_1$ for a single spin molecule and a multitraiplet excitation gap $\Delta_2$, that is, those that give the partition function $Z = 1 + 3 \exp(-\Delta_1/kT) + 3 \exp(-\Delta_2/kT)$, the elastic modulus $C_T(T)$ in the molecular-spin system is
written in the mean-field approximation as \(^\text{14}\):

\[
C_\Gamma(T) = C_\Gamma^0(T) - C_{1,\Gamma}^2 N \frac{\chi_\Gamma(T)}{1 - K_\Gamma \chi_\Gamma(T)},
\]

where \(C_\Gamma^0(T)\) is the background elastic constant, \(N\) the density of spin molecules, \(C_{1,\Gamma} = \frac{\partial \Delta_1}{\partial \epsilon_{\text{spin}}}\) the coupling constant for a single spin molecule measuring the strain (\(\epsilon_{\text{spin}}\)) dependence of the excitation gap \(\Delta_1\), \(K_\Gamma\) the interspin-molecule interaction, and \(\chi_\Gamma(T)\) the strain susceptibility of a single spin molecule. Equation (3) expresses the temperature dependence of the strain susceptibility, which is the response function to an applied strain, and its form is analogous to the mean-field expression for the magnetic susceptibility, which is the response function of the magnetization to an applied magnetic field.

We now give a quantitative analysis of the experimental \(C_\Gamma(T)\) in ZnFe\(_2\)O\(_4\) using Eq. (3) with the assumption of the excitations of the AF dodecamers in the Fe\(^{3+}\) pyrochlore lattice shown in Fig. 1(e)\(^\text{15}\) an excitation gap \(\Delta_1\) for a single AF dodecamer and a multi-AF-dodecamer excitation gap \(\Delta_2\). Fits of Eq. (3) to the experimental data \(C_\Gamma(T)\) for ZnFe\(_2\)O\(_4\) are given in Fig. 3(a)-(c) as solid curves. The fits correspond to temperatures below 50 K. Here, the value of the density of AF dodecamers \(N = 1.73 \times 10^{27} \text{ m}^{-3}\) in Eq. (3) is fixed in a first approximation\(^\text{15}\). Although the background \(C_\Gamma^0(T)\) in Eq. (3) generally exhibits hardening with decreasing temperature,\(^\text{15}\) we here assume that \(C_\Gamma^0(T)\) is constant because, for ZnFe\(_2\)O\(_4\), the hardening below \(\sim 50\) K is negligibly small compared with the softening-with-minimum in \(C_\Gamma(T)\), as indicated in Fig. 2(a)-(c). Given the values of the fitting parameters listed in Table I, the fits to Eq. (3) are in excellent agreement with the experimental data of ZnFe\(_2\)O\(_4\) (Fig. 3(a)-(c)), reproducing the characteristic minimum in \(C_\Gamma(T)\). The \(K_\Gamma\) values are negative for all the elastic modes, indicating that the inter-AF-dodecamer interaction is antiferrodistortive. Moreover, the coupling constant \(G_{2,\Gamma} = \frac{\partial \Delta_2}{\partial \epsilon_{\text{spin}}}\) is larger than \(G_{1,\Gamma} = \frac{\partial \Delta_1}{\partial \epsilon_{\text{spin}}}\), indicating that the higher excitations \(\Delta_2\) couple to the lattice deformations more strongly than the lowest excitations \(\Delta_1\). Both the coupling constants \(G_{1,\Gamma}\) and \(G_{2,\Gamma}\) exhibit the largest values for the trigonal shear modulus \(C_{44}(T)\) of the three elastic moduli, and hence is compatible with the trigonal symmetry of the AF dodecamer. The insets to Fig. 3(a)-(c) depict the symmetry of the spin molecules as well as the propagation \(k\) and polarization \(u\) of the sound waves. The AF-dodecamer excitations should couple more sensitively to the trigonal lattice deformations generated by sound waves with \(k \parallel [110]\) and \(u \parallel [001]\) (Fig. 3(c)).

In Table I, the values of the fit parameters of Eq. (3) for the experimental \(C_{44}(T)\) of ACr\(_2\)O\(_4\) (\(A = \text{Mg and Zn}\)) are also listed for comparison.\(^\text{16}\) For ACr\(_2\)O\(_4\), the value of \(N = 3.45 \times 10^{27} \text{ m}^{-3}\) in Eq. (3) is fixed in a first approximation, where the spin molecule is assumed to be the Cr\(^{3+}\) AF hexamer (Fig. 1(d))\(^\text{11,13}\). As described before in conjunction with Fig. 2(a)-(c) and Fig. 2(f), softening in \(C_\Gamma(T)\) begins to occur below \(\sim 50\) K or \(\sim 80\) K in ZnFe\(_2\)O\(_4\) but above 300 K in MgCr\(_2\)O\(_4\). In accordance with Eq. (3), this difference in the temperature region between ZnFe\(_2\)O\(_4\) and ACr\(_2\)O\(_4\) arises from the difference

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**TABLE I**: Values of the fitting parameters in Eq. (3) for the experimental \(C_\Gamma(T)\) for ZnFe\(_2\)O\(_4\) and ACr\(_2\)O\(_4\) (\(A = \text{Mg and Zn}\)). The fitted curves for ZnFe\(_2\)O\(_4\) are plotted as solid curves in Fig. 3(a)-(c). The values for ACr\(_2\)O\(_4\) are from Ref.\(^\text{16}\).

| \(\Delta_1\) (K) | \(G_{1,\Gamma}\) (K) | \(\Delta_2\) (K) | \(G_{2,\Gamma}\) (K) | \(K_\Gamma\) (K) |
|------------------|-------------------|-----------------|------------------|----------------|
| ZnFe\(_2\)O\(_4\) | \(C_{11}\) | 890 | 2630 | -6 |
| \(\text{(Dodec.)}\) | \(C_t\) | 5 | 980 | 15 | 2910 | -10 |
| \(C_{44}\) | 1350 | 3570 | -6 |
| MgCr\(_2\)O\(_4\) | \(C_{44}\) | 39 | 3600 | 136 | 10200 | -19 |
| ZnCr\(_2\)O\(_4\) | \(C_{44}\) | 34 | 3160 | 111 | 9290 | -19 |
in the coupling strength between the lattice deformation and molecular-spin excitations. From Table I, the coupling constants $G_{1,\Gamma}$ and $G_{2,\Gamma}$ for $C_{44}(T)$ of ZnFe$_2$O$_4$ are $\sim$2 to $\sim$3 times smaller than those of ACr$_2$O$_4$, respectively. Additionally, along with Fig. 3(a)-(c) and Fig. 2(f), $C_\Gamma(T)$ for ZnFe$_2$O$_4$ exhibits a minimum at $\sim$4 K or $\sim$6 K, which is lower than the minimum point of $\sim$50 K in $C_{44}(T)$ of MgCr$_2$O$_4$. From Eq. (3), this difference in the minimum point between ZnFe$_2$O$_4$ and ACr$_2$O$_4$ arises from the difference in the magnitudes of the gaps in the molecular-spin excitations.

From the listing in Table I, the gaps $\Delta_1$ and $\Delta_2$ in ZnFe$_2$O$_4$ are $\sim$7 to $\sim$9 times smaller than those corresponding to ACr$_2$O$_4$. The smaller values for ZnFe$_2$O$_4$ indicate the formation of spin molecules by the effectively weaker exchange interactions involving the different exchanges for $J_1$ and $J_2$. This formation in ZnFe$_2$O$_4$ results from frustration in the effectively weaker exchange interactions, which is compatible with the smaller magnitude of the Weiss temperature in ZnFe$_2$O$_4$ ($|\Theta_W| \approx 120$ K) than ACr$_2$O$_4$ ($|\Theta_W| \approx 390$ K). Thus, for ZnFe$_2$O$_4$, it is suggested that the spin-lattice coupling cannot produce the spin-JT transition because the strength of the exchange interactions is not large enough to overcome the cost in elastic energy involved in the lattice deformation, resulting therefore in the emergence of solely the dynamical molecular-spin state.

For ZnFe$_2$O$_4$, the magnitudes of $\Delta_1$ and $\Delta_2$ listed in Table I are comparable to those of the inter-spin-molecule interaction $K_\Gamma$, which is in contrast to the stronger magnitudes of $\Delta_1$ and $\Delta_2$ than $K_\Gamma$ in ACr$_2$O$_4$. The comparable magnitudes of $\Delta_1$, $\Delta_2$, and $K_\Gamma$ in ZnFe$_2$O$_4$ are compatible with the presence of a very weak dispersive feature of the molecular-spin excitations in the INS spectra suggesting the presence of the inter-spin-molecule correlations. Also, taking into account the smaller values of $\Delta_1$ and $\Delta_2$ in ZnFe$_2$O$_4$ than ACr$_2$O$_4$, the comparable magnitudes of $\Delta_1$, $\Delta_2$, and $K_\Gamma$ in ZnFe$_2$O$_4$ imply that the formation of the spin molecules is not so robust compared with that for ACr$_2$O$_4$. This might be a result of frustration occurring in the effectively weaker exchange interactions.

We finally note that the recent high-resolution INS experiments in MgCr$_2$O$_4$ revealed the presence of multiple modes associated with finite-energy molecular-spin excitations, where it is suggested that not only the ground state but also the excited states are highly frustrated in this compound. Hence, although with the assumption of two gaps $\Delta_1$ and $\Delta_2$ Eq. (3) gives successful agreement with experimental data for $C_\Gamma(T)$ in both ZnFe$_2$O$_4$ and ACr$_2$O$_4$, it is expected that the level schemes of the spin molecules in these compounds consist of not only the excited levels of $\Delta_1$ and $\Delta_2$ but also higher excited levels. Similar to MgCr$_2$O$_4$, the future high-resolution INS experiments in ZnFe$_2$O$_4$ are expected to reveal the complex and exotic molecular-spin excitations.

IV. SUMMARY

In summary, we performed ultrasound velocity measurements on a single crystal of the spin-frustrated ferrous spinel ZnFe$_2$O$_4$. All the symmetrically-independent elastic moduli in the cubic crystal exhibited anomalies, which evidence the presence of molecular-spin excitations. Additionally, the present study also revealed that the elastic anomalies driven by the spin-JT phase transition and its precursor are absent in ZnFe$_2$O$_4$, suggesting that the spin-lattice coupling cannot play a role in releasing frustrations in this compound. Further experimental and theoretical studies are indispensable if the dynamical molecular-spin state in ZnFe$_2$O$_4$, which is expected to govern the magnetic properties, is to be understood.

V. ACKNOWLEDGMENTS

This work was partly supported by a Grant-in-Aid for Scientific Research (C) (Grant No. 25400348) from MEXT of Japan, and by Nihon University College of Science and Technology Grants-in-Aid for Project Research.

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