Ultrasonics in the two-dimensional dimer spin system YbAl$_3$C$_3$

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Abstract. We report on results of ultrasonic investigations of the two-dimensional dimer spin system YbAl$_3$C$_3$ in magnetic fields ($H$) up to 10 T and for temperatures ($T$) down to 0.5 K. A magnetic field induces a pronounced minimum in the elastic constants far below the transition temperature of 80 K. The minimum disappears immediately on increasing $T$ and becomes undetectable above around 2 K. We propose the two-dimensional dimer spin model with the energy difference of $\Delta=15$ K between the singlet ground state and the triplet excited one. We discuss the mechanism how to form the two-dimensional dimer spin system in YbAl$_3$C$_3$ where Yb ions have a high degree of geometrical frustration. Our observations show an important role of an exchange striction coupling in YbAl$_3$C$_3$ in the ordered state below 80 K.

1. Introduction

Ytterbium (Yb)-based intermetallic compounds have generated considerable interest in recent years. Especially, the discovery of a huge low-temperature Sommerfeld coefficient $\gamma$ that exceeds 8 J/mol K$^2$ has motivated the search for new intriguing quantum phenomena and anomalous metallic materials in Yb-based systems.[1,2] On the other hand, a study of the geometrical frustration effects in the $f$-electron systems has attracted much attention in several compounds, such as the pyrochlore oxides $RTi_2O_7$, a quasi-Kagome antiferromagnet YbAgGe and $RB_4$ with the Shastry-Sutherland lattice, where $R$ denotes rare-earth elements.[3-5] YbAl$_3$C$_3$ has a hexagonal unit cell with $c/a \sim 5.04$, and each layer, which is composed of Yb, Al, or C, stacks along the $c$ axis. The effective paramagnetic moment follows well the expected value for the Yb$^{3+}$ ion, indicating that Yb ion is well localized in YbAl$_3$C$_3$. Accordingly, the network of Yb atoms can be regarded to be a two-dimensional triangular lattice as follows. The $J$-multiplet of the Yb$^{3+}$ ion is split into four Kramers doublets by the hexagonal crystalline electric field (CEF) effects. The first excited state is most likely to lie at 20 meV from the ground state. [6] Thus, the ground state Kramers doublet governs the low temperature properties of the system. However, this split of CEF ground state is quite unusual since no magnetic long-range order can be observed down to 20 mK in spite of the magnetic Kramers doublet with a large negative $\theta_p$ of -94 K.[7]. Generally, the rare-earth intermetallic compounds having the Kramers doublet ground state undergoes a magnetic ordering in order to release the magnetic entropy of the ground state. However, the geometrical frustration effect possibly suppresses a long range magnetic ordering.

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in this system, again although $\theta_p$ indicates a quite large antiferromagnetic interaction. This situation is likely to lead a dimerization of Yb$^{3+}$ ions at low temperatures.

In spite of such a geometrical frustration system, YbAl$_3$C$_3$ exhibits a clear phase transition at 80 K. This ordering was initially suggested to be a possible antiferroquadrupolar (AFQ) ordering. However, the discovery of the same clear phase transition in the reference system LuAl$_3$C$_3$ made us change the scenario what occurs at 80 K. Alternatively, a simple structural phase transition scenario was proposed. The magnitude is, however, extremely small, and the lattice parameters do not change within the experimental accuracy. Atoms are only slightly displaced. In this paper, we present the evidence for the existence of the spin gap state and a clear elastic anomaly associated with the singlet-triplet excitations in the isolated Yb$^{3+}$ dimer in YbAl$_3$C$_3$ at low temperatures.

2. Experiment

Polycrystalline sample of YbAl$_3$C$_3$ was prepared by using an encapsulated alumina crucible sealed in a quartz ampoule filled with argon atmosphere. The obtained sample was confirmed to be in a unique phase by the powder X-ray diffraction measurement. The details are described elsewhere. YbAl$_3$C crystallizes in ScAl$_3$C$_3$-type structure (space group P63/mmc, $a = 3.394$, $c = 17.11\ \text{Å}$)

The sound velocity, as the elastic constant was measured by an ultrasonic apparatus based on a phase comparison method at temperatures down to 0.5 K in magnetic field up to 10 T. Plates of LiNbO$_3$ were used for the piezoelectric transducer. The fundamental resonance frequency of LiNbO$_3$ transducer is 10 - 30 MHz. The transducer was glued on the parallel planes of the sample by an elastic polymer Thiokol. The absolute value of the sound velocity was obtained by measuring the delay time between the ultrasonic echo signals with an accuracy of a few percent. The elastic constant was calculated as $C = \rho v^2$ by using the sound velocity $v$ and the density $\rho$ of the crystal. The lattice constant of YbAl$_3$C$_3$ at room temperature was used for the estimation of the density $\rho = 2.82 \text{ g/cm}^3$.

![Figure 1. Magnetic field dependence of a longitudinal elastic constant $C_L$ at selected temperatures of YbAl$_3$C$_3$. Red circles and blue solid lines denote experimental data and calculated results based on the formula (2).](image-url)
Magnetic field (T)
1.5 K
0.5 K
  calculation
  experiment
1 %

Figure 2. Magnetic field dependence of a transverse elastic constant \( C_T \) at selected temperatures of YbAl\(_3\)C\(_3\). Red circles and blue solid lines denote experimental data and calculated results based on the formula (2).

Table 1. The absolute values of a longitudinal elastic constant \( C_L \), transverse one \( C_T \), Lamé's constants \( \lambda \), bulk modulus \( K \), Young's modulus \( Y \) and Poisson's ratio \( \sigma \) of YbAl\(_3\)C\(_3\) at 4.2 K.

|        | \( C_L \) (GPa) | \( C_T=\mu \) (GPa) | \( \lambda \) (GPa) | \( K \) (GPa) | \( Y \) (GPa) | \( \sigma \) |
|--------|-----------------|----------------------|---------------------|--------------|--------------|------------|
| (4.2 K)| 29.0            | 1.69                 | 25.7                | 26.8         | 4.97         | 0.469      |
| (77 K) | 27.9            | 1.64                 | 24.6                | 25.7         | 4.81         | 0.468      |

3. Experimental results
We measured the longitudinal as well as transverse ultrasonic velocity. We used 10 - 30 MHz for the measurement of \( C_L \) and 5 - 15 MHz for \( C_T \). Red circles in Fig. 1 show the magnetic field \( (H) \) dependence of longitudinal mode measured on the polycrystalline YbAl\(_3\)C\(_3\) sample at selected temperatures of 0.5 K and 1.5 K. A pronounced minimum was observed in the \( C_L - H \) curves. The minimum becomes sharper with decreasing \( T \). The same behavior was also recognized in the \( H \) dependence of transverse mode as shown in Fig. 2. Namely, a pronounced minimum was observed in the \( C_T - H \) curves. It is noteworthy that the minimum was only observed below 1.5 K. It is undetectable anymore when increasing \( T \) at 2.0 K, although the data is not shown here. The reason why the elastic anomaly is observed only at low temperatures will be discussed below in detail, which is typical for low-dimensional spin systems. The absolute values of elastic constants and calculated Lamé's constants \( \lambda \) and \( \mu \) at 4.2 K are listed in Table I. The Lamé's constants \( \lambda \) and \( \mu \) are described by \( C_L \) and \( C_T \) as \( C_L=\lambda + 2\mu \) and \( C_T=\mu \). Thus, Bulk modulus \( K \), Young’s modulus \( Y \) and Poisson’s ratio \( \sigma \) are calculated by \( \sigma = \lambda / 2(\lambda + \mu) \), \( Y = \mu (3\lambda + 2\mu) / (\lambda + \mu) \) and \( K = \lambda + 2\mu / 3 \).

4. Discussions
Let us discuss the minimum in the \( C_L \)- and \( C_T \) - \( H \) curves. As for the elastic anomalies investigated previously in SrCu\(_2\)(BO\(_3\))\(_2\) and RB\(_4\), an exchange striction effect is possibly responsible for these phenomena.[5, 10-12] This exchange striction coupling arises from the strain dependence of the exchange interaction. The single dimer susceptibility \( \chi^0 \) may be calculated by considering the splitting of a dimer into a singlet ground state and an excited state at energy
\[ \Delta \] In such a system, the free energy for this system can be written by

\[
F = \frac{1}{2} C_0 (\varepsilon_1^2 + \varepsilon_2^2) - \frac{1}{\beta} \ln Z
\]

with the partition function \( Z = \exp\{-\beta(\Delta + d_1 \varepsilon_1 + d_2 \varepsilon_2)\} + \exp\{-\beta(\Delta + d_1 \varepsilon_1 + d_2 \varepsilon_2 - g \mu_B H)\} + \exp\{-\beta(\Delta + d_1 \varepsilon_1 + d_2 \varepsilon_2)\} \), where \( C_0 \) is the background elastic constant, \( \varepsilon_1 \) and \( \varepsilon_2 \) are the desired elastic strains coupled the singlet ground state and excited one with the constants \( d_1 \) and \( d_2 \), respectively. \( \beta \) is \( 1/k_B T \), where \( k_B \) is the Boltzmann constant.

In this model the single dimer coupling constant \( d \) between the elastic strain and the dimer e.g., exchange striction coupling plays a crucial role. Finally, we obtain for the isothermal elastic constant by making \( \varepsilon_1 \) and \( \varepsilon_2 \) nearly equal to the same value of \( \varepsilon \) and sufficiently close to zero as following,

\[
C_T(T) = \frac{\partial^2 F}{\partial \varepsilon^2} = C_0 - 4d^2 \beta \frac{\exp(-\beta \Delta) [1 + 2 \cosh(g \mu_B H \beta)]}{[1 + \exp(-\beta \Delta)][1 + 2 \cosh(g \mu_B H \beta)]^2}
\]

This expression gives a reasonable fit to the experimental results as seen from Figs. 1 and 2 with a value of the energy difference of \( \Delta = 15 \) K, \( g \)-value of 2.13 and the coupling constant \( d \) of 15 K and 3 K for the longitudinal and transverse modes, respectively. The value of \( \Delta = 15 \) K is in good agreement with that of previous reports such as the specific heat, magnetic susceptibility and INS measurements.[6, 8] The exchange striction coupling describes reasonably the \( H \) dependence of the elastic constant. The spin of Yb\(^{+3} \) ions that are responsible for the magnetism are grouped in dimers within planes of the hexagonal YbAl\(_3\)C\(_3\) unit cell.

5. Summary

In summary, we present ultrasonic measurements in the two-dimensional dimer spin system YbAl\(_3\)C\(_3\) in magnetic fields up to 10 T and for temperatures down to 0.5 K. From anomalies in the \( C_L \)- and \( C_T \)- \( H \) curves we suggest the dimer spin system of Yb\(^{+3} \) with the energy difference of \( \Delta = 15 \) K between the singlet ground state and the triplet excited one. The present study indicates that the exchange striction coupling should be taken into account for model describing the properties of YbAl\(_3\)C\(_3\) in the ordered state below 80 K, especially at low temperatures.

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