Elastic properties of SmRu$_4$P$_{12}$ in pulsed magnetic fields

P Sun$^1$, Y Nakanishi$^1$, T Kono$^2$, H Sugawara$^3$, D Kikuchi$^4$, H Sato$^4$ and M Yoshizawa$^1$

$^1$ Graduate School of Engineering, Iwate University, Morioka 020-8551, Japan
$^2$ Superconductivity Research Laboratory, ESTEC, Morioka Laboratory for Applied Superconductivity Technology, Morioka 020-0852, Japan
$^3$ Faculty of Integrated Arts and Science, The University of Tokushima, Tokushima 770-8502, Japan
$^4$ Graduate School of Science, Tokyo Metropolitan University, Hachioji 192-0397, Japan

E-mail: psun@iwate-u.ac.jp (P. Sun)

Abstract. A setup for ultrasonic measurement in pulsed magnetic fields was established in this work. We first introduce the basic ideas of this measurement. Using this setup we measured the longitudinal elastic constant $C_{11}$ and the corresponding ultrasound attenuation $\beta_{11}$ up to 28 T for the filled skutterudite compound SmRu$_4$P$_{12}$. At 14 K, $C_{11}$ shows steep increase while $\beta_{11}$ a large peak at around 8 T, indicating the magnetic ordering of this compound. Anomalies were also observed at 4.2 K, however, it is not clear if these correspond to phase transitions.

1. Introduction

Ultrasonic technique that measures elastic constant and ultrasound attenuation can provide helpful information on the electron-lattice interaction in materials [1]. Many of such experiments have to be performed in magnetic fields for some reasons, e.g., to gain knowledge on an ordering parameter or to detect phase transition in magnetic fields. In this work we intend to perform such measurements for the filled skutterudite compound SmRu$_4$P$_{12}$. This compound is attracting lots of attentions because of its two mystery successive transitions. One is at $T_{MI} = 16.5$ K known as a metal-insulator (MI) transition, the subsequent one is at about $T_N \sim 14$ K known as an antiferro-magnetic one [2]. The magnetic ordering at $T_N$ is very obscure at zero magnetic field while is enhanced in magnetic field [3, 4]. In contrast with the iso-structural MI transition material PrRu$_4$P$_{12}$ [5], no structural distortion is found at the MI transition for SmRu$_4$P$_{12}$. On the other hand, the $B$-$T$ phase diagram of SmRu$_4$P$_{12}$ [6, 7] has a strong resemblance with that of the typical antiferro-quadrupolar (AFQ) ordering material CeB$_6$ [8]. Therefore, multipolar interaction including quadrupole and octupole is believed to dominate the transitions [9]. However, the ordering parameter remains to be an enigma.

The first problem in this work is that we have to perform ultrasonic measurements in higher magnetic field up to 30 T, because the critical field increases rapidly by varying temperature [6, 7]. However, most of the currently available superconducting magnets have limited maximum field less than 20 T. In this work, we first establish a new setup for performing ultrasonic measurements in pulsed high magnetic fields, then measure the longitudinal elastic constant $C_{11}$ and the corresponding ultrasound attenuation $\beta_{11}$ for SmRu$_4$P$_{12}$.
2. Experimental technique

A phase sensitive detection technique was used in the ultrasonic measurements. This technique usually measures the relative ultrasound velocity $\Delta \upsilon/\upsilon$ by detecting the relative shift of ultrasound frequency $\Delta f/f$ [1]. Here, the frequency $f$ is shifted by a feedback loop when $\upsilon$ (then the phase $\phi$) changes as a function of external parameter (e.g., temperature or magnetic field). However, the measurements in pulsed magnetic fields have to be performed using a fixed frequency. This is because the feedback loop used to adapt the frequency is too slow in comparison with a magnetic field pulse. In the case of a fixed frequency for pulsed magnetic field, one has to detect the ultrasonic echo in two 90° shifted channels. The main principles of this technique have ever been described previously [10]. However, no phase discrepancy from 90° of the two channels were considered there. We will describe the process in detail considering the probable phase discrepancy.

![Figure 1](image1.png)

**Figure 1.** Frequency scan of $V_1$ and $V_2$ of the two perpendicular channels in 0 T. Both of the two voltages oscillate with frequency in sine curve. The $f_p$ indicates a frequency cycle. The inset presents the Lissajous curve of $V_1$ vs. $V_2$, where $\phi_1$ and $\phi_2$ indicate the phase discrepancies from 90°.

The signals $V_1$ and $V_2$ of the two perpendicular channels on the $n$-th echo is generally described as $V_n = A \sin[\phi_n + (2\pi f/f_p)]$. This feature is shown in figure 1 as the frequency scan of $V_1$ and $V_2$. Here $A$ denotes echo amplitude, $\phi_n$ the phase, $f_p$ one frequency cycle as indicated in the figure. Based on this relation, the phase $\phi_n$ can be expressed as $\phi_n = \arctan(V_1/V_2)$; a phase shift $\Delta \phi_n$ can be described with a frequency shift as $\Delta f/f = -(f_p/2\pi f)\Delta \phi_n$. Therefore, following the basic idea for phase sensitive technique as mentioned above, the relative change of ultrasound velocity and amplitude at a fixed frequency can be calculated by the equations $\Delta \upsilon/\upsilon = -(f_p/2\pi f)\arctan(V_1/V_2)$ and $A = (V_1^2 + V_2^2)^{1/2}$, respectively.

![Figure 2](image2.png)

**Figure 2.** The $V_1$, $V_2$, and the magnetic field $B$ as a function of time recorded in one cycle of the pulsed magnetic field at 14 K.

However, the two channels are usually not well separated into perpendicular phases by a quadrature hybrid. As shown in figure 1 (inset), the Lissajous curve is not a regular circle. In this case, the phase must be modified by estimating the discrepancy $\phi_1$ of $V_1$ and $\phi_2$ of $V_2$, as shown in the inset. For convenience, we suppose $\phi_1 = -\phi_2$ in our measurements, whose value is found to be less than 5° in general. After calibrating the phase, the relative ultrasound velocity and amplitude should be modified as follows.

$$\frac{\Delta \upsilon}{\upsilon} = - \frac{f_p}{2\pi f} \arctan \left[ \frac{V_1 \cos \phi_2 - V_2 \sin \phi_1}{V_2 \cos \phi_1 + V_1 \sin \phi_2} \right].$$  \hspace{1cm} (1)
A = \frac{1}{|\cos(\phi_1 - \phi_2)|}\left[V_1^2 + V_2^2 - 2V_1V_2\sin(\phi_1 - \phi_2)\right]^\frac{1}{2}. \quad (2)

We analyzed our data using Eq. 1 and Eq. 2. Relative elastic constant \( \Delta C/C \) is twice of \( \Delta \nu/\nu \) considering the general formula \( C = \rho \nu^2 \). Ultrasound attenuation \( \beta \) is obtained by the equation

\[ \beta = -\frac{20}{T_0} \log A. \]

The position of the gated echo, which is around 10 \( \mu s \) in our measurements.

Pulsed magnetic fields were generated by feeding a pulsed current to a solenoid coil. The pulsed current was supplied by a capacitor bank with a capacitance of 8 mF. The duration of the magnetic field pulse is 14 ms, including a half cycle of reversed field. The single crystal of \( \text{SmRu}_4\text{P}_{12} \) employed here was prepared by tin flux method and have a dimension of about 1 x 1 x 1 mm\(^3\). A couple of LiNbO\(_3\) piezoelectric transducers are bonded to the two parallel faces of the single crystal sample in the measurement. We used a typical repetition rate of about 20 kHz for the incidence ultrasound. That is to say, in a half cycle (7 ms) of the pulsed field, about 140 effective points can be obtained. Longitudinal ultrasound mode \( C_{11} \) with polarization and propagation vectors of sound wave parallel to the \( <001> \) direction was measured. In figure 2, we show a set of raw data recorded in one pulse cycle. The symmetrical variation of the voltages with respect to magnetic fields indicates the intrinsic changes of elastic constant and attenuation.

3. Results and discussions

![Figure 3. Longitudinal elastic constant \( C_{11} \) and the corresponding ultrasound attenuation \( \beta_{11} \) measured at 14 K as a function of magnetic field.](image)

Figure 3 shows the longitudinal elastic constant \( C_{11} \) and the corresponding ultrasound attenuation \( \beta_{11} \) as a function of magnetic field measured at 14 K. Both \( C_{11} \) and \( \beta_{11} \) are calculated results by the raw data shown in figure 2. At 8 T, a sharp peak in \( \beta_{11} \) and a steep increase in \( C_{11} \) (hardening) are observed. Because this temperature is at the boundary of the probable multipolar ordering phase (phase II) and the magnetic ordering phase (phase III), these anomalies can be reasonably attributed to the II-III phase transition in \( \text{SmRu}_4\text{P}_{12} \). \( \Delta C_{11}/C_{11} \) has a change of about 0.4\%, which is common for a magnetic ordering however is very small compared with a typical quadrupolar ordering. On the other hand, the sharp peak in attenuation is noticeable. Such an enhanced peak of ultrasound attenuation as a function of magnetic field is seldom reported for a magnetic phase transition. One more significant feature of the \( \beta_{11} \) is, it has enhanced values in phase III and is weaken considerably in phase II. This is not well understood at the moment.

Figure 4 shows \( C_{11} \) and \( \beta_{11} \) measured at 4.2 K. The relative change of \( C_{11} \) is rather small in comparison with that at 14 K but has a clear structure. Two obscure anomalies can be confirmed at about 16 T and 25 T on both \( C_{11} \) and \( \beta_{11} \). A crystal electric field (CEF) scheme or
phase transitions may be the origin. Specific heat measurements have suggested a CEF scheme consisting of a \( \Gamma_{67} \) quartet ground state and a \( \Gamma_5 \) excited doublet at about 60 K [6]. However, this CEF scheme may be largely modulated by the magnetic ordering and the magnetic fields. Thus it is not clear whether this structure of \( C_{11} \) at 4.2 K reflects a modulated CEF scheme or phase transitions.

4. Summary
In summary, we established a new setup for ultrasonic measurements in pulsed magnetic fields. Elastic constant and ultrasound attenuation were measured in pulsed magnetic fields up to 28 T. Distinct anomalies were observed both in ultrasound attenuation and in elastic constant on the II-III phase boundary at 14 K for SmRu\textsubscript{4}P\textsubscript{12}. At 4.2 K anomalies were also observed, which are not understood at the moment. Measurements at other temperatures and for other elastic modes of \( C_{44} \) and \( (C_{11}-C_{12})/2 \) are in progress.

Acknowledgements
This work is supported by Grant-in-Aid for Scientific Research Priority Area, Skutterudite (no. 15072202) of the Ministry of Education, Culture, Sports, Science and Technology of Japan.

References
[1] Thalmeier P and Lüthi B, in Handbook on the Physics and Chemistry of Rare Earths, edited by Gschneidner K A et al. (North-Holland, Amsterdam, 1991), Vol 14 p 225
[2] Sekine C, Uchiumi T, Shirotani I and Yagi T, in Science and Technology of High Pressure, edited by Manghani M H et al. (Universities Press, Hyderabad, India, 2000) p 826
[3] Matsuura K, Doi Y, Wakeshima M, Hinaitsu T, Amitsuka H, Shimaya Y, Giri R, Sekine C and Shirotani I, 2005 J. Phys. Soc. Jpn. 74 1030
[4] Yoshizawa M, Nakanishi Y, Kumagai T, Oikawa M, Sekine C and Shirotani I, 2004 J. Phy. Soc. Jpn. 73 315
[5] Lee C H, Matsuhara H, Yamamoto A, Ohta T, Takazawa H, Ueno K, Sekine C, Shirotani I and Hirayama T, 2001 J. Phys.: Condens. Matter 13 L45
[6] Matsuura K, Hinaitsu T, Sekine C, Togashi T, Maki H, Shirotani I, Kitazawa H, Takamasu T and Kido G, 2002 J. Phys. Soc. Jpn. Suppl. 71 237
[7] Sekine C, Shirotani I, Matsuura K, Haen P, Brion S D, Chouteau G, Suzuki H and Kitazawa H, 2003 Acta Physica Polonica B 34 983
[8] Effantin J M, Rossat-Mignod J, Burlet P, Bartholin H, Kunii S and Kasuya T, 1985 J. Magn. Magn. Mater. 47-48 145
[9] Yoshizawa M, Nakanishi Y, Oikawa M, Sekine C, Shirotani I, Saha S R, Sugawara H and Sato H, 2005 J. Phy. Soc. Jpn. 74 2141
[10] Wolf B, Bruls G, Kouroudis I, Finsterbusch D, Sieling M, Schmidt S, Palme W and Lüthi B 1998 Physica B 246-247 179