Microscopic Investigation of Field Sensitive Electronic State in β-US_2: ^{33}S-NMR Study

Sakai, H., Higa, N., Hattori, T., Tokunaga, Y. and Kambe, S. (ASRC, JAEA); Reyes, A.P. (NHMFL), Rosa, P.F.S., Thompson, J.D., Ronning, F. and Bauer, E.D. (LANL)

Introduction

Uranium disulfide β-US_2 is a paramagnet and narrow-gap semiconductor with an energy band gap of 90 K [1] in the low temperatures, while it is a semimetal above 80 K with an extremely low density of carriers. Another characteristic feature in β-US_2 is the strong response of the electrical resistivity to magnetic field. The magnetoresistance is huge but very anisotropic. Interestingly, if pulsed external field up to 55 T is applied, the semiconducting behavior is completely suppressed [2]. To investigate this intriguing material microscopically, we began to perform ^{33}S-NMR research in 2014. Even though 33-Sulfur (^{33}S) has a nuclear spin of I=3/2, it is usually NMR-inert due to its low nuclear gyromagnetic ratio (γ_n=3.2654 MHz/T) and very low natural abundance of 0.76%. In this respect, it is a great challenge to utilize ^{33}S-NMR in condensed matter physics research. We have started the project in JAEA to grow 50%-^{33}S enriched crystals of β-US_2 for NMR purposes. Since the samples cannot be imported from Japan, we also grew the 50%-^{33}S enriched samples in Los Alamos National Laboratory (LANL). The LANL crystals were successfully grown and we found that it appears to be semimetallic all the way down to lowest temperatures, as shown in Fig. 1. The difference may be caused by an alternate transport agent (NH_4Cl vs Br_2), which may cause a small number of carriers to be doped or may introduce additional in-gap states. It is also interesting to identify the source of this variation in conductivity.

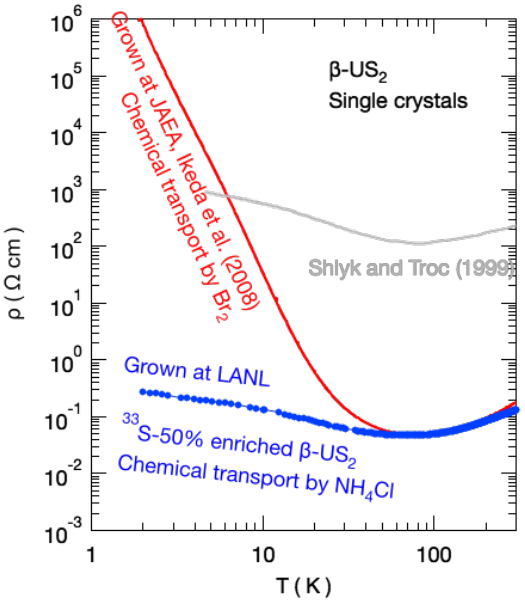

Fig. 1 Temperature dependence of electrical resistivity for the single crystals of β-US_2 grown at JAEA and LANL. The values in Ref. 3 are plotted.

Results and Discussion

There are two different crystallographic S sites in β-US_2. When external field is applied, each of the two S sites is further differentiated with respect to local field directions. The ^{33}S-NMR spectrum shows multiple transitions as shown in Fig. 3(c). The nuclear quadrupolar parameters can be derived, which represent the electric field gradients on each ^{33}S site, as well as the values of Knight shifts. The temperature dependence of the Knight shifts has been confirmed to be very anisotropic. If the temperature dependence is compared with that of the magnetic susceptibility χ, the hyperfine coupling constants can be drawn. These analyses in JAEA and NHMFL are ongoing. Nuclear relaxation rates $1/T_1$ have been measured under H_0=7, 12, and 17 T. The $(T_1T)^{-1}$, corresponding to the ***q***-summed imaginary part of dynamical electronic susceptibility, is found to respond strongly to external fields. Above 100 K, the system seems to have strong excitation due to localized *f* moments, and the $(T_1T)^{-1}$ has no significant increase by external fields. Below 100 K, however, the response of $(T_1T)^{-1}$ becomes prominent, although it sharply decreases with an energy gap of ~20 K.

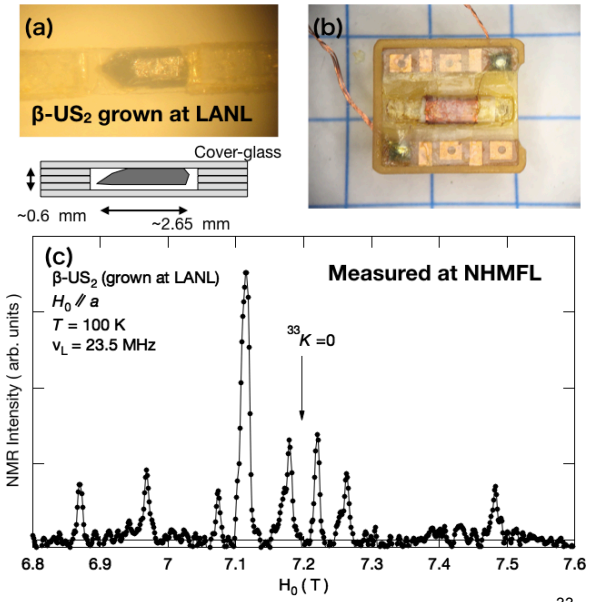

Fig. 2 (a) Photo of a single crystal of 50%-^{33}S enriched β-US_2. (b) Photo of NMR coil setup. (c) Field-swept ^{33}S-NMR spectrum with a constant frequency of ν_L=23.5 MHz for a crystal of β-US_2 with H_0 ∥ *a* axis, taken with the NHMFL spectrometer.

Acknowledgements

The National High Magnetic Field Laboratory is supported by the National Science Foundation through NSF/DMR-1157490/1644779 and the State of Florida. A part of this work was supported by JSPS KAKENHI Grant Number JP16KK0106.

References

[1] Ikeda, S. *et al.*, J. Phys. Soc. Jpn. **78**, 114704-1–10 (2009).
[2] Sugiyama, S. *et al.*, J. Phys. Soc. Jpn. Suppl. A **80**, SA104-1–3 (2011).
[3] Shlyk, L. and Troc, R., Physica B **262**, 90–97 (1999).