Two-gap superconductivity in $Lu_2Fe_3Si_5$: A transverse-field muon spin rotation study

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The superconducting properties of $Lu_2Fe_3Si_5$ with $T_c = 6.1$ K have been investigated using low-temperature transverse-field muon spin rotation (μSR) and specific-heat measurements. The magnetic penetration depth at zero temperature, $\lambda(0)$, is 353(1) nm. The temperature dependence of the magnetic penetration depth $\lambda(T)$ is consistent with a two-gap $s+s$-wave model. Low-temperature specific-heat measurements on the same sample also show evidence of two distinct superconducting gaps.

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I. INTRODUCTION

The discovery of superconductivity in MgB_2 with a $T_c \sim 39$ K (Ref. 1) has generated a great deal of interest in superconducting materials containing light elements such as B, C, and Si. Among these materials, the ternary iron silicide superconductors $R_2Fe_3Si_5$, with $R =$ Lu, Y, or Sc, are particularly noteworthy due to the presence of iron.[2,3] $Lu_2Fe_3Si_5$ is the most interesting of the ternary iron silicide superconductors because of its high superconducting transition temperature ($T_c = 6.1$ K), large upper critical field ($\mu_0 H_{c2} = 6$ T),[2,4] and unconventional superconducting properties. Recently, a detailed study of the low-temperature specific heat on a single crystal of $Lu_2Fe_3Si_5$ revealed a two-gap superconductivity similar to that seen in MgB_2.[5] $Lu_2Fe_3Si_5$ has a tetragonal $Sc_2Fe_3Si_5$-type structure (space group $P4/mnc$) consisting of quasi-one-dimensional iron chains along the c axis and quasi-two-dimensional iron squares parallel to the basal plane.[6]

Muon spin rotation (μSR) is an ideal probe with which to study the mixed state of type-II superconductors, as it provides microscopic information of the local field distribution within the bulk of the sample. It has often been used to measure the temperature dependence of the London magnetic penetration depth λ in the vortex state.[7,8] The temperature and field dependence of λ can provide information on the nature of the superconducting gap.

Here we have investigated the unusual superconducting properties of $Lu_2Fe_3Si_5$ by carrying out low-temperature μSR measurements on a polycrystalline sample. We show that the temperature dependence of λ can be well described using a two-gap $s+s$-wave model. The magnetic penetration depth at $T = 0$ K is estimated to be $\lambda(0) = 353(1)$ nm. We also study the low-temperature specific heat of $Lu_2Fe_3Si_5$ in order to support the validity of the two-gap model. We compare these results with published data for the $R_2Fe_3Si_5$ system.

II. EXPERIMENTAL DETAILS

A polycrystalline sample of $Lu_2Fe_3Si_5$ was prepared by melting a stoichiometric mixture of lutetium shot (99.99%), iron granules (99.999%), and silicon pieces (99.99%) in an arc furnace under an argon atmosphere. The as-cast sample was poorly superconducting with a $T_c = 4.8$ K and a broad transition. In order to improve these characteristics, it is essential to anneal the as-cast samples at high temperature for a long period of time.[5,9] The as-cast sample was sealed in a quartz tube under a partial pressure of argon. The sample was then heated at a rate of 200°C/h to 800°C, held at this temperature for 48 h, then heated at the same rate to 1100°C and held at this temperature for 72 h. The sample was then cooled at 200°C/h to 800°C, maintained at this temperature for 72 h, and then finally cooled to room temperature. dc magnetic susceptibility versus temperature (T) measurements were performed using a Quantum Design Magnetic Property Measurement System (MPMS) magnetometer. The temperature dependence of the magnetic susceptibility shows that the annealed sample of $Lu_2Fe_3Si_5$ has a transition temperature, T_c (onset), of 6.1 K (see Fig. 1). It is the superconducting properties of these annealed samples that are discussed below. Low-temperature specific-heat measurements were carried out using a two-τ relaxation method in a Quantum Design Physical Property Measurement System (PPMS) equipped with a ^{3}He insert.

The μSR experiments were performed on the MuSR spectrometer of the ISIS pulsed muon facility. The transverse-field (TF)-μSR experiments were conducted with applied fields between 5 and 60 mT, which ensured that the sample was in the mixed state. The magnetic field was either applied above the superconducting transition and the sample then cooled to base temperature [field cooled (FC)], or the sample was first cooled to base temperature and then the field was applied [zero-field cooled (ZFC)]. The MuSR spectrometer comprises 64 detectors. In software, each detector is normalized for the muon decay and the data is then reduced to two orthogonal components which are then fitted simultaneously.

The sample was mounted on a silver plate with a circular area of ~700 mm^2 and a small amount of diluted GE varnish was added to aid thermal contact. The sample and mount were then inserted into an Oxford Instruments ^{3}He sorbtion cryostat. Any silver exposed to the muon beam gives a nondecaying sine wave.

III. RESULTS AND DISCUSSION

TF-μSR precession signals above and below $T_c = 6.1$ K are shown in Fig. 2. Above the superconducting transition, i.e., in the normal state, the signal decays very slowly, but the decay is relatively fast in the superconducting state due to the inhomogeneous field distribution from the flux-line lattice.

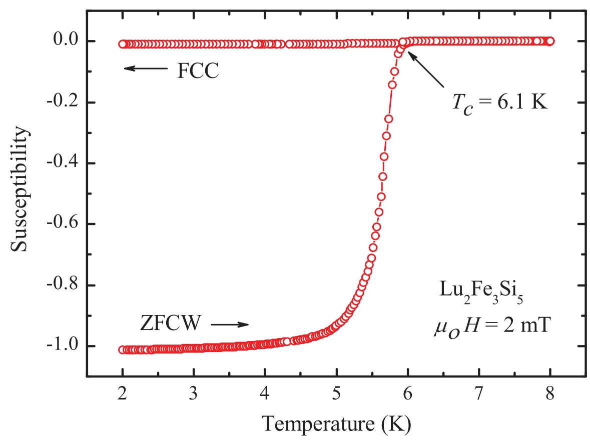

FIG. 1. (Color online) The temperature dependence of the dc magnetic susceptibility of $Lu_2Fe_3Si_5$ measured using both zero-field-cooled warming (ZFCW) and field-cooled cooling (FCC). The diamagnetic susceptibility shows a T_c onset of 6.1 K.

We can model these inhomogeneous field distributions using an oscillatory decaying Gaussian function,

$$G_X(t) = A_0 \exp(-\Lambda t)\exp(-\sigma^2 t^2/2)\cos(\omega_1 t + \phi) + A_1 \cos(\omega_2 t + \phi), \quad (1)$$

where ω_1 and ω_2 are the frequencies of the muon precession signal and background signal, respectively, ϕ is the initial phase offset, and σ is the Gaussian muon spin relaxation rate. σ can also be defined as $\sigma = (\sigma_{sc}^2 + \sigma_{nm}^2)^{1/2}$, where σ_{sc} is the superconducting contribution to the relaxation rate and σ_{nm} is the nuclear magnetic dipolar contribution which is assumed to be constant over the entire temperature range. Figure 3(a) shows the temperature dependence of σ_{sc} obtained in an applied TF of 0.03 T. Figure 3(b) presents the magnetic-field dependence of σ_{sc} collected at different temperatures below the superconducting transition. A deviation in the field dependence of σ_{sc} is observed at 40 mT in the data collected at 0.3 K. A small deviation of σ_{sc} is also present at the same field in the data collected at 2 K, whereas it is constant above 2 K.

The temperature dependence of the London magnetic penetration depth, $\lambda(T)$, is coupled with the superconducting Gaussian muon spin depolarization rate $\sigma_{sc}(T)$ by the equation

$$\frac{2\sigma_{sc}^2(T)}{\gamma_\mu^2} = 0.00371\frac{\Phi_0^2}{\lambda^4(T)}, \quad (2)$$

where $\gamma_\mu/2\pi = 135.5$ MHz/T is the muon gyromagnetic ratio and $\Phi_0 = 2.068 \times 10^{-15}$ Wb is the magnetic-flux quantum.[7,8] $\lambda(T)$ can be calculated within the local London approximation[10,11] by the following expression:

$$\frac{\lambda^{-2}(T,\Delta_{0,i})}{\lambda^{-2}(0,\Delta_{0,i})} = 1 + \frac{1}{\pi}\int_0^{2\pi}\int_{\Delta(T,\varphi)}^{\infty}\left(\frac{\partial f}{\partial E}\right)\frac{E\,dE\,d\varphi}{\sqrt{E^2 - \Delta_i(T,\varphi)^2}}, \quad (3)$$

where $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function, φ is the angle along the Fermi surface, and $\Delta_i(T,\varphi) = \Delta_{0,i}\delta(T/T_c)g(\varphi)$. The temperature dependence of the gap is approximated by the expression $\delta(T/T_c) = \tanh\{1.82[1.018(T_c/T - 1)]^{0.51}\}$ while $g(\varphi)$ describes the angular dependence of the gap and is replaced by 1 for both an s-wave and an $s+s$-wave gap, and $|\cos(2\varphi)|$ for a d-wave gap.[12,13]

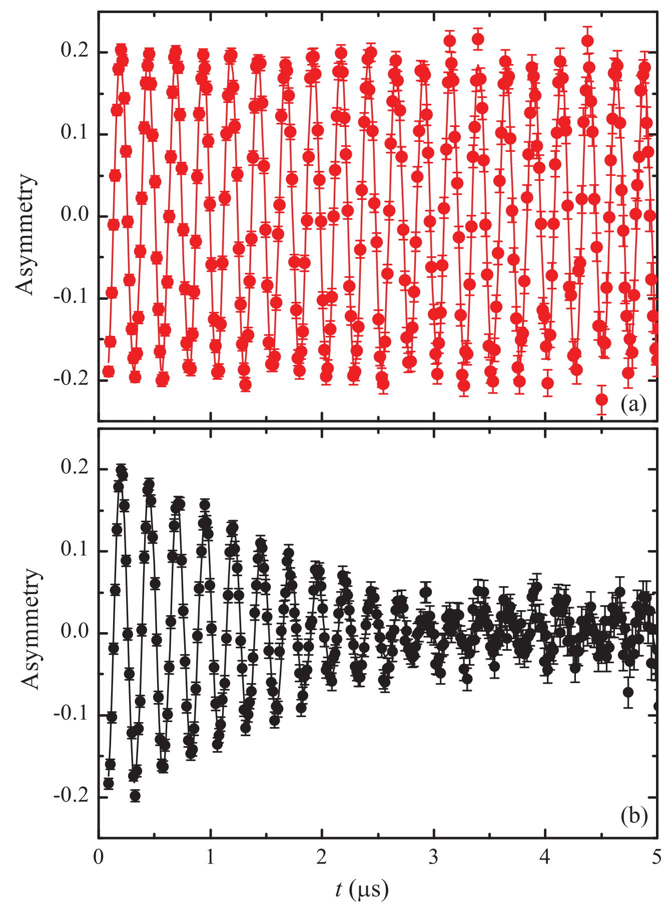

FIG. 2. (Color online) The transverse-field muon time spectra (one component) for $Lu_2Fe_3Si_5$ collected (a) at $T = 6.5$ K and (b) at $T = 0.3$ K in a magnetic field $\mu_0 H = 30$ mT.

The temperature dependence of the penetration depth can then be fitted using either a single gap or a two-gap model which are structured on the basis of the α model,[14,15]

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \omega_1\frac{\lambda^{-2}(T,\Delta_{0,1})}{\lambda^{-2}(0,\Delta_{0,1})} + \omega_2\frac{\lambda^{-2}(T,\Delta_{0,2})}{\lambda^{-2}(0,\Delta_{0,2})}, \quad (4)$$

where $\lambda^{-2}(0)$ is the penetration depth at zero temperature, $\Delta_{0,i}$ is the value of the ith ($i = 1$ or 2) superconducting gap at $T = 0$ K, and ω_i is a weighting factor with $\omega_1 + \omega_2 = 1$.

Fits to the data using the three different models are shown in Fig. 4. The fits appear to rule out the s-wave and d-wave models as possible descriptions for $Lu_2Fe_3Si_5$ as the χ^2 values for these models are 33.92 and 15.91, respectively. The two-gap $s+s$-wave model gives a good fit to the data with a χ^2 of 1.94. The two-gap $s+s$-wave model gives $\Delta_{0,1}/k_B T_c = 1.76(6)$ and $\Delta_{0,2}/k_B T_c = 0.40(1)$ with $\omega_1 = 0.35(1)$. The ratio of the larger to the smaller gap, $\frac{\Delta_{0,1}}{\Delta_{0,2}} \approx 4.40$, which is consistent with the value 5 obtained by low-temperature specific-heat measurement,[5] and the value 3.44 obtained by penetration depth measurement using the tunnel-diode resonator technique[16] on a single crystal of $Lu_2Fe_3Si_5$. The magnetic penetration depth at $T = 0$ K is

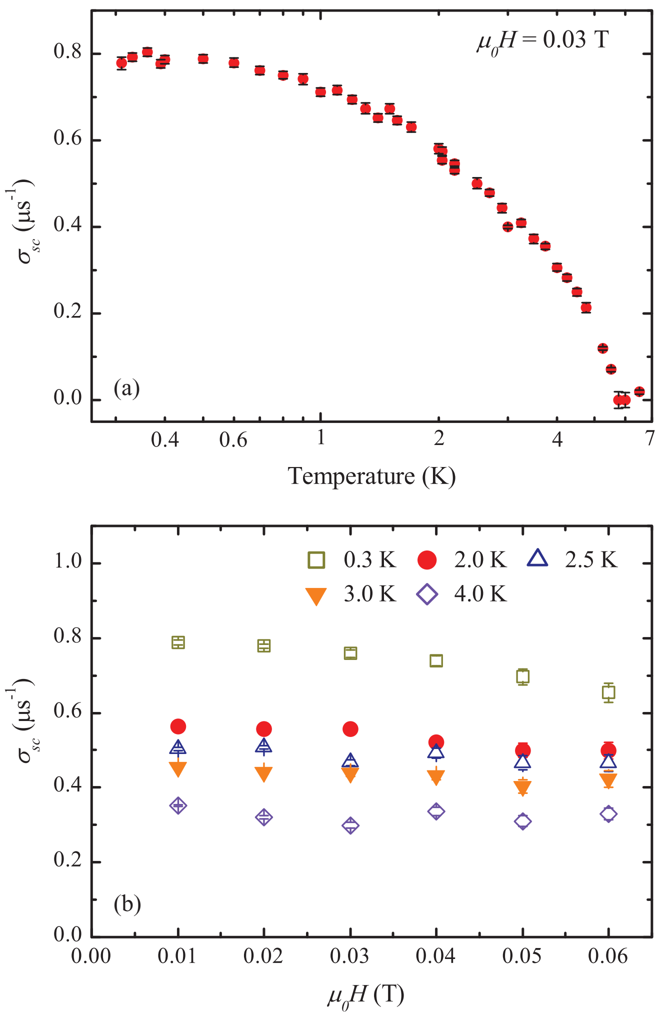

FIG. 3. (Color online) (a) The temperature dependence (on a log scale) of the superconducting muon spin depolarization rate σ_{sc} collected in an applied magnetic field $\mu_0 H = 30$ mT. (b) Superconducting Gaussian depolarization rate σ_{sc} versus applied magnetic field for $Lu_2Fe_3Si_5$ collected below T_c at 0.3, 2.0, 2.5, 3.0, and 4.0 K.

found to be λ(0) = 353(1) nm. The in-plane penetration depth is 200 nm, obtained by tunnel-diode resonator technique.[16]

We have also performed low-temperature specific-heat measurements on $Lu_2Fe_3Si_5$ which support the assertion that $Lu_2Fe_3Si_5$ is a two-gap superconductor. Figure 5 shows the specific heat divided by temperature (C/T) as a function of T^2 for the same polycrystalline sample of $Lu_2Fe_3Si_5$ used for the μSR study. A pronounced jump in the specific heat is observed at 6.1 K which indicates that the sample exhibits bulk superconductivity. The normal-state heat capacity has been fitted up to 12 K by $C = \gamma T + \beta T^3 + \alpha T^5$, where γT is the electronic contribution and $\beta T^3 + \alpha T^5$ represents the lattice contribution to the specific heat. We obtained fitted parameters $\gamma = 24.9$ mJ/mol K^2, $\beta = 0.247$ mJ/mol K^4, and $\alpha = 5.38 \times 10^{-4}$ mJ/mol K^6, which are consistent with the reported values for both polycrystalline[17–19] and single-crystal samples.[5] We observed a sizable residual specific-heat coefficient, $\gamma_\circ = 7.21$ mJ/mol K^2, at $T = 0$ K. Interestingly, a finite residual specific-heat coefficient has also been observed in a polycrystalline sample of the same system[19] whereas it is absent in data for a single crystal.[5] A similar effect has also been reported in $Ba_{0.6}K_{0.4}Fe_2As_2$ ($\gamma_\circ =$ 7.7 mJ/mol K^2), $Ba(Fe_{1-x}Co_x)_2As_2$ ($\gamma_\circ = 3.0$ mJ/mol K^2) and $Ba(Fe_{0.92}Co_{0.08})_2As_2$ ($\gamma_\circ = 3.7$ mJ/mol K^2).[20–22] Possible explanations for this residual specific-heat coefficient involve pair breaking effects of an unconventional superconductor,[23] spin-glass behavior, or crystallographic defects.[22] Given the metallurgy of our polycrystalline sample and the dramatic effects that annealing has on the electronic properties, we suggest that crystallographic defects are the most likely cause of the residual specific-heat coefficient in heat-capacity data.

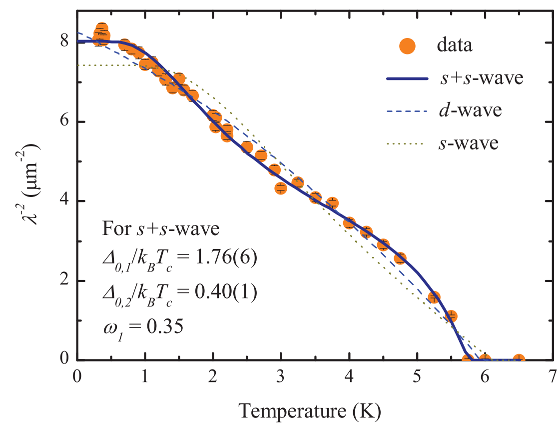

FIG. 4. (Color online) The temperature dependence of the London penetration depth as a function of temperature for $Lu_2Fe_3Si_5$. The solid line is a two-gap $s + s$-wave fit to the data while the dashed and dotted lines represent the d-wave and s-wave fits, respectively.

We have calculated the normalized electronic specific heat $C_e/\gamma T$ by subtracting the lattice contribution from the total heat capacity and then renormalizing by $(\gamma - \gamma_\circ)$. For more details, see Refs. 22 and 24. Figure 6 shows the temperature dependence of the normalized electronic specific heat, $C_e/\gamma T$, for $Lu_2Fe_3Si_5$ as a function of T/T_c. We find two clear anomalies in the data which show that $Lu_2Fe_3Si_5$ has two energy gaps. A large jump appears at T_c and a smaller one at $T_c/5$. The value of $C_e/\gamma T$ at T_c is found to be 1.13(1) meV, which is much smaller than the BCS value of

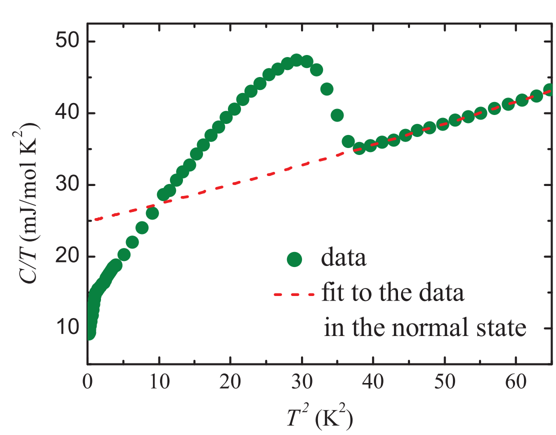

FIG. 5. (Color online) The specific heat divided by temperature (C/T) as a function of T^2 for $Lu_2Fe_3Si_5$. The dashed line shows the fit to the data in the normal state.

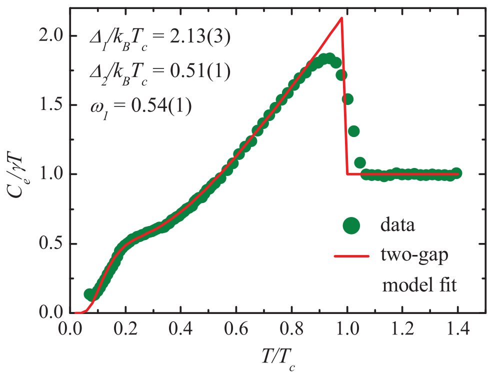

FIG. 6. (Color online) The temperature dependence of the normalized electronic specific heat as a function of T/T_c for $Lu_2Fe_3Si_5$. The solid line is a two-gap fit to the data.

1.43 meV but consistent with the value of 1.05 meV measured on a single crystal[5] and also agrees well with the reported values for polycrystalline samples.[17–19] To perform a two-gap fit to the $C_e/\gamma T$ data in the superconducting state, we use the BCS expressions for the normalized entropy S and the specific heat,

$$\frac{S}{\gamma_n T_c} = -\frac{6}{\pi^2}\frac{\Delta_0}{k_B T_c}\int_0^\infty [f\ln f + (1-f)\ln(1-f)]dy, \quad (5)$$

$$\frac{C}{\gamma_n T_c} = t\frac{d(S/\gamma_n T_c)}{dt}, \quad (6)$$

where $t = T/T_c$, $E = [\epsilon^2 + \Delta^2(t)]$, and $y = \epsilon/\Delta$. The temperature dependence of the energy gap varies as $\Delta(t) = \Delta_0\delta(t)$, where $\delta(t)$ is the normalized BCS gap.[25] The solid line in Fig. 6 is a two-gap fit to the data, with $\Delta_1/k_B T_c = 2.13(3)$ and $\Delta_2/k_B T_c = 0.51(1)$. The weighting factor, $\omega_1 = 0.54$, is slightly larger than the value obtained from fits to the μSR data. The good agreement between the experimental heat-capacity data and the two-gap model argues in favor of the presence of two distinct superconducting gaps in $Lu_2Fe_3Si_5$. The ratio of the two gaps, $\frac{\Delta_1}{\Delta_2} = 4.18$, is close to the 4.40 obtained from the μSR measurements on the same sample and is also consistent with the published data on $Lu_2Fe_3Si_5$.[5,17–19] The quantitative discrepancy between the gap magnitudes calculated from the μSR and the specific-heat studies may be due to a coupling between the two bands that cannot be accounted for using the simple two-gap model adopted here. Kogan *et al.*[26] recently presented a self-consistent calculation of the heat capacity of a two-band s-wave superconductor in the clean limit with one interband and two intraband pairing potentials. This model was tested with experimental data collected on the well-known two-band superconductors MgB_2 and V_3Si. A more sophisticated analysis using such a self-consistent model may be required to fully understand the two-band superconducting behavior in $Lu_2Fe_3Si_5$.

IV. SUMMARY

We have performed a μSR study on a polycrystalline sample of $Lu_2Fe_3Si_5$. The temperature dependence of the magnetic penetration depth data was fitted with three different models. A two-gap $s + s$-wave model provides the best fit to the data. Low-temperature specific-heat measurements on the same sample confirm the presence of two distinct superconducting gaps. These results are consistent with other reported data for this system.[5,16–19]

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