Multigap superconductivity in the charge density wave superconductor $LaPt_2Si_2$

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(Received 28 February 2018; revised manuscript received 1 May 2018; published 18 May 2018)

The superconducting gap structure of a charge density wave (CDW) superconductor $LaPt_2Si_2$ ($T_c = 1.6$ K) having a quasi-two-dimensional crystal structure has been investigated using muon spin rotation/relaxation (μSR) measurements in transverse field (TF), zero field (ZF), and longitudinal field (LF) geometries. Rigorous analysis of TF-μSR spectra in the superconducting state corroborates that the temperature dependence of the effective penetration depth, λ_L, derived from muon spin depolarization, fits to a two gap s wave model (i.e., $s+s$ wave) suggesting that the Fermi surface contains two gaps of different magnitude rather than an isotropic gap expected for a conventional s wave superconductor. On the other hand, ZF μSR data do not show any significant change in muon spin relaxation rate above and below the superconducting transition temperature indicating the fact that time-reversal symmetry is preserved in the superconducting state of this material.

DOI: 10.1103/PhysRevB.97.184509

I. INTRODUCTION

BCS theory [1–3], which explains superconductivity in conventional systems, fails to unfold the mystery of witnessing superconductivity in some materials which form a new class of superconductors (SC), collectively classified as unconventional SC. This encompasses a variety of materials which includes cuprate, heavy-fermion superconductor, pnictide, etc. [4–8]. Unlike conventional SC where pairing is mediated by lattice vibrations or phonons, in unconventional SC, fluctuations of the order parameters play a crucial role in the formation of a superconducting ground state. Hence, the search for unconventional SC and understanding their pairing mechanism has become an intensely studied active research area for the past few decades. In this quest, discovery of superconductivity by suppressing spin density wave (SDW) ordering in Fe-based pnictides has received considerable attention [7–10]. Both spin fluctuations and density fluctuations (associated with structural transition) are believed to be important in governing superconductivity in the system. Very recently, charge density wave (CDW) systems which can be recognized as nonmagnetic analog of Fe-based pnictides, has gained significant research interest as the fluctuations associated with CDW are believed to be a key factor in inducing superconductivity in the system [11–15].

Recently, RPt_2Si_2 (R = La, Pr) system has attracted an intensive research interest as it exhibits strong interplay between CDW and superconductivity [15–19]. RPt_2Si_2 crystallizes in primitive tetragonal $CaBe_2Ge_2$ type structure (space group $P4/nmm$) [15–17] having a close resemblance to the $ThCr_2Si_2$ type structure found in pnictide and heavy fermion SC. However, the striking difference between these two structures is that the former one lacks inversion symmetry in the crystal structure which contains two inequivalent [Ge1-Be2-Ge1] and [Ge2-Be1-Ge2] layers with Ca atom (or R atom for RPt_2Si_2) being sandwiched between them [20]. In this context it is to be mentioned that other than RPt_2Si_2, $SrPt_2As_2$ is another example which crystalizes in this crystal structure and exhibits coexistence of superconductivity and CDW [11,12,21]. Moreover, this special feature in crystal structure is reminiscent of noncentrosymmetric SC [22–26] where the lack of inversion symmetry results in nonuniform lattice potential which in turn creates an asymmetric spin orbit coupling allowing mixing of pairing symmetry between a spin singlet and a spin triplet cooper pairs. Mixing of spin singlet and triplet pairing makes these noncentrosymmetric SC more likely to exhibit time reversal symmetry (TRS) breaking and the physics of the system can be modified by this broken symmetry. TRS breaking is rare and has only been observed directly in a few unconventional SC, e.g., Sr_2RuO_4 [27], UPt_3 [28], $(U,Th)Be_{13}$ [29], $(Pr,La)(Os,Ru)_4Sb_{12}$ [30], $PrPt_4Ge_{12}$ [31], $LaNiC_2$ [24], $LaNiGa_2$ [32], Re_6Zr [33], and $(Lu,Y)_5Rh_6Sn_{18}$ [34,35]. Zero field muon spin relaxation (ZF − μSR) is a powerful tool to search for very weak TRS breaking fields or spontaneous internal field below $T_{\mathbf{c}}$. The presence of such low internal field limits the pairing symmetry and mechanism responsible for unconventional superconductivity. However, it is well established that this mixing of spin states does not always indicate TRS breaking [36,37].

In this framework, $LaPt_2Si_2$ turns out to be quite an interesting system hosting a number of unusual phenomena such as lack of inversion symmetry in crystal structure, CDW

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transition, structural phase transition from tetragonal to orthorhombic structure and superconductivity [15–17,38]. Through small angle electron diffraction study [15], CDW wave vector has been confirmed to be $(n/3, 0, 0)$, where n (= 1, 2) is the order of reflection, which requires tripling of the unit cell below T_{CDW}. Furthermore, theoretical prediction of coexistence of CDW and superconductivity in $LaPt_2Si_2$ by Kim *et al.* [39] has been confirmed experimentally in our earlier reports [16,17]. On the other hand, electronic structure calculation predicts quasi-two-dimensional nature of the Fermi surface [40] similar to that seen for iron pnictides. These observations conjointly hint towards an exotic origin of superconductivity in the system. However, the superconducting gap structure, which is intimately related to the superconducting mechanism, remains unexplored until now. It requires microscopic techniques in order to have a proper understanding of the superconducting phase which emerges in the presence of a competing CDW phase. This motivates us to perform μSR experiments in the superconducting state to unveil the gap structure in $LaPt_2Si_2$. μSR is a method to resolve the type of pairing symmetry in superconductors [41]. In case of a type-II SC, the mixed or vortex state gives rise to a spatial distribution of local magnetic fields influencing the μSR signal through a relaxation of the muon spin polarization. In this paper, we present the results of our detailed μSR investigation performed on $LaPt_2Si_2$ compound. Our results manifest the existence of multigap superconductivity in this system.

II. EXPERIMENTAL DETAILS

A high quality polycrystalline sample of $LaPt_2Si_2$ was prepared by arc melting the constituent elements taken in stoichiometric amount on a water cooled copper hearth in argon atmosphere, followed by annealing at 1000°C for a week. The detailed procedure of sample preparation can be found in Ref. [16]. Phase purity of the polycrystalline sample was checked by powder x-ray diffraction (XRD) using Cu-K_α radiation. XRD pattern obtained at room temperature was analyzed by Rietveld refinement using FullProf software [42]. The detail of sample characterization has been provided in the Supplemental Material [43]. The μSR experiments were performed in the MUSR spectrometer at ISIS pulsed muon facility of the Rutherford Appleton Laboratory, United Kingdom [44]. The μSR measurements had been carried out in transverse-field (TF), zero-field (ZF), and longitudinal-field (LF) configurations. The powdered sample was mounted on a high purity (99.999%) silver plate using diluted GE varnish and covered with a thin Ag foil which was cooled down to 50 mK in a commercial dilution refrigerator (ICE). 100% spin-polarized muon pulses were implanted into the sample and positrons from the resulting decay were collected in the detectors. TF-μSR experiments were carried out in the superconducting mixed state under different applied magnetic fields ranging from 100 G to 300 G. TF-μSR measurements were performed in the field cooled mode in which the magnetic fields were applied above the superconducting transition temperature and the sample was then cooled down to 50 mK. For ZF-μSR measurements, the sample was cooled down to base temperature in true zero field. In both cases, data were collected while warming the sample. μSR data were analyzed using the free software package WIMDA [45].

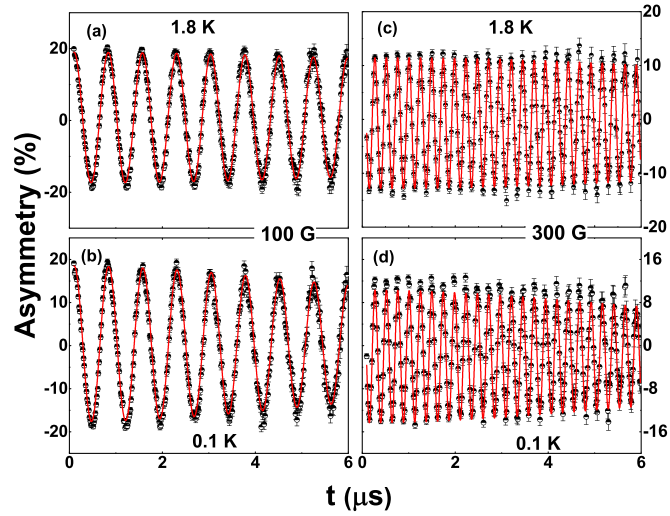

FIG. 1. Transverse field μSR spectra (one component) for $LaPt_2Si_2$ obtained at $T = 1.8$ K and at $T = 0.1$ K in an applied magnetic field of 100 G [see (a),(b)] and 300 G [see (c),(d)] for field-cooled (FC) state. Solid red lines represent the fits to the observed spectra with Eq. (1).

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) and 1(c) and 1(d) show the TF-μSR precession signals (above and below T_c) obtained in FC condition under an applied field of 100 G and 300 G, respectively. It can be seen from Fig. 1 that below T_c, the μSR precession signal decays with time in both the cases caused by the inhomogeneous field distribution of the flux-lattice emphasizing the fact that the sample is indeed in the superconducting mixed state. Observed TF-μSR asymmetry spectra can be best fitted with an oscillatory decaying Gaussian function which is given by

$$G_{TF}(t) = A_0 \cos(2\pi\nu_1 t + \phi)\exp\left(-\frac{\sigma_{\mathrm{tot}}^2 t^2}{2}\right) + A_{BG}\cos(2\pi\nu_2 t + \phi), \quad (1)$$

where ν_1 and ν_2 represent the frequencies of muon precession signal originating from the superconducting fraction of the sample and the background due to sample holder, respectively. A_0 and A_{BG} are the muon initial asymmetries associated with the sample and background, respectively, σ_{tot} is the total sample relaxation rate, and ϕ is the initial phase offset. Fitting of the observed spectra with Eq. (1) is presented by the solid red line in Figs. 1(a)–1(d). Below T_c, the values of internal field in the superconducting state are lower than the applied field due to diamagnetic shift, which is expected for type-II superconductors [46]. Inset of Fig. 2(a) represents the temperature dependence of the internal field for an applied field of 100 G. Now considering the information related to the superconducting gap structure, the first term in Eq. (1) is most important, as below T_c it gives the total sample relaxation rate σ_{tot} which contains contributions from the vortex lattice (σ_{sc})

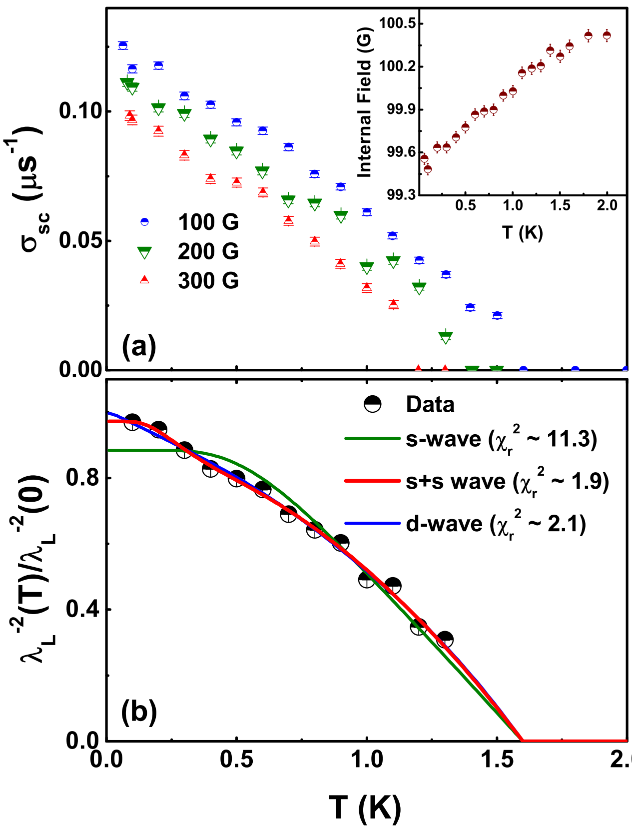

FIG. 2. (a) Temperature dependence of the muon depolarization rate σ_{sc} of $LaPt_2Si_2$ measured under applied magnetic field of 100 G, 200 G, and 300 G in field cooled (FC) condition. Inset: Temperature dependence of the internal field for an applied field of 100 G. (b)Variation of normalized inverse magnetic penetration depth, $\lambda_L^{-2}(T)/\lambda_L^{-2}(0)$, as a function of temperature. The lines are fit to the data using an isotropic s-wave model, linear combination of two s waves (i.e., $s+s$ wave) model, and d wave model with line nodes.

and nuclear dipole moments (σ_{nm}), the latter is expected to be constant over the entire temperature range (i.e., above and below T_c). σ_{tot} is related to σ_{sc} and σ_{nm} by the relation $\sigma_{\text{tot}} = \sqrt{\sigma_{sc}^2 + \sigma_{nm}^2}$. Thus, the contribution due to the vortex lattice, σ_{sc}, was obtained by quadratically subtracting the background nuclear dipolar relaxation rate obtained from the fitting of the spectra measured above T_c.

We obtained the magnetic field and temperature dependence of $\sigma_{sc}(T,H)$ by fitting Eq. (1) to the μSR time dependent asymmetry spectra. Figure 2(a) depicts the temperature dependence of σ_{sc} for three different applied fields. After that, we used the numerical Ginzburg-Landau model developed by Brandt [47],

$$\sigma_{sc}[\mu s^{-1}] = 4.83 \times 10^4 (1 - H/H_{c2}) \times [1 + 1.21\sqrt{(1 - H/H_{c2})^3}]\lambda_L^{-2}[nm] \quad (2)$$

to fit the field-dependent depolarization rate $\sigma_{sc}(H)$ and estimate two important superconducting order parameters, i.e., the London penetration depth λ_L and the upper critical field H_{c2}. This model presumes that λ_L is field independent. Now σ_{sc} is directly related to the magnetic penetration depth (λ_L) which is associated with the superconducting gap structure. Therefore, σ_{sc} can be modeled with the superconducting gap

TABLE I. Superconducting parameters obtained by fitting μSR data with different models.

Model	Gap ($\Delta_0/k_B T_c$)	$\lambda_L(0)$ (nm)	$n_s \times 10^{27}$ (m^{-3})	χ_r^2
s wave	1.523	292.86	4.99	11.3
d wave	2.278	275.95	5.63	2.1
$s+s$ wave	1.846	279.36	5.49	1.9
	0.475			

by the relation [48]

$$\frac{\sigma_{sc}(T)}{\sigma_{sc}(0)} = \frac{\lambda_L^{-2}(T)}{\lambda_L^{-2}(0)} = 1 + 2\left\langle \int_{\Delta}^{\infty} \int_0^{2\pi} \frac{\delta f}{\delta E} \frac{E dE d\varphi}{\sqrt{E^2 - \Delta_k^2}} \right\rangle_{FS}, \quad (3)$$

where f is the Fermi function given by $f = [1 + \exp(E/k_B T)]^{-1}$, the brackets $\langle\rangle_{FS}$ signify the averaging over the Fermi surface, and Δ represents the superconducting gap. This gap Δ which is a function of temperature and the azimuthal angle (φ) along the Fermi surface can be described as $\Delta(T,\varphi) = \Delta_0 \delta(T/T_c) g(\varphi)$. Here, the temperature dependence of the superconducting gap is approximated by the relation [49] $\delta(T/T_c) = \tanh\{1.82[1.018 * (T_c/T - 1)]^{0.51}\}$. The spacial dependence $g(\varphi)$ takes the value 1 for s wave and $|\cos(2\varphi)|$ for d wave model with line nodes [48–50]. We have analyzed $\lambda_L^{-2}(T)/\lambda_L^{-2}(0)$ data estimated from the TF-μSR data analysis of 100, 200, and 300 G as shown in Fig. 2(b) using Eq. (3). We have considered three models in our analysis: an isotropics (s wave) gap model, a combination of two s waves with different gaps (i.e., $s+s$ wave model), and d wave model with line nodes.

In Table I, we have summarized superconducting gap parameter values obtained after fitting with different models. It can be seen from Fig. 2(b) that both the d wave model and $s+s$ wave model replicate the observed data quite well.

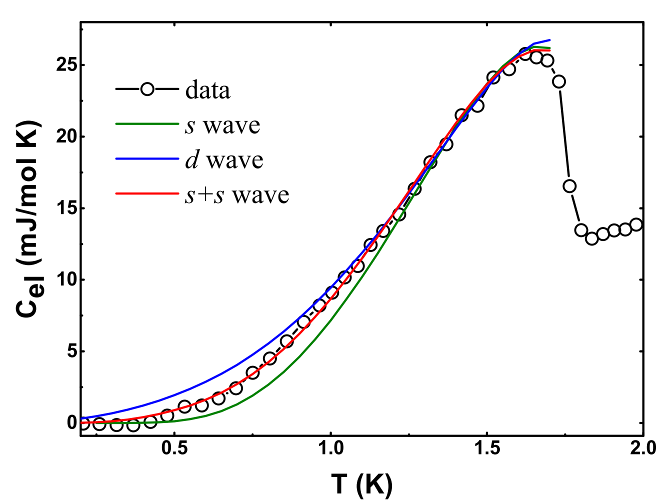

FIG. 3. Variation of the electronic part of specific heat measured on single crystalline sample. Solid lines correspond to the fitting of experimentally obtained data with different models as described in the text.

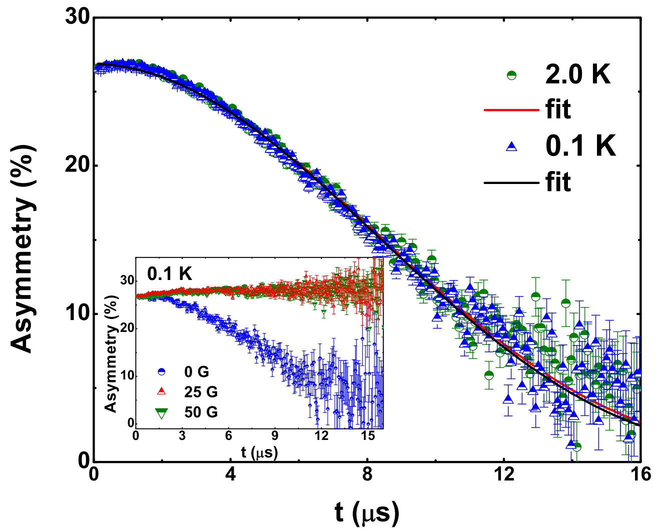

FIG. 4. ZF μSR spectra of $LaPt_2Si_2$ recorded at $T = 0.1$ K and 2 K. Solid line represents fitting of the observed spectra with damped KT function (see text). Inset: Comparison of LF spectra measured for an applied field of 0, 25 G, and 50 G at 0.1 K.

But this apparent dilemma can be resolved by having a close look at the goodness of the fit which suggests that the $s+s$ model gives the lowest value of reduced χ_r^2 (defined as $\chi_r^2 = \chi^2/(n\text{-}m)$; where n is the number of data points and m is the number of fit parameters) indicating the best fit (for s wave, d wave, and $s+s$ wave models, χ_r^2 values are found out to be 11.3, 2.1, and 1.9, respectively) to the observed data. Furthermore, in order to uphold this conclusion drawn from μSR investigation, we have performed a detailed analysis of the electronic part of specific heat (see Supplemental Material [43] for detail analysis). Within the formulation of BCS theory [51,52], the entropy S_{sc} can be expressed by the relation $S_{sc} = -\frac{3\gamma_n}{\kappa_B \pi^3}\int_0^{2\pi}\int_0^{\infty}[(1-f)ln(1-f)+f ln f]d\varepsilon d\varphi$, where γ_n is the normal state Sommerfeld coefficient, κ_B is the Boltzmann constant, and f is the Fermi function defined as above with $E = \sqrt{\varepsilon^2 + \Delta^2(\varphi,T)}$. The temperature dependence of the gap function was defined using well established α model [43,53,54]. Now S_{sc} is related to the electronic part of the specific heat C_{el} with the relation $C_{el} = T(\frac{\delta S_{sc}}{\delta T})$. Thus, applying the same methodology as described above we can test different models for C_{el}. Figure 3 represents C_{el} as a function of temperature obtained on a single crystalline sample [17] and fitting of the same with different models (s wave, d wave with line node, and $s+s$ wave models). It clearly suggests that the $s+s$ wave model gives the best fit of the data which is in accordance with the μSR analysis. Hence, analysis of specific heat and μSR data conjointly hint towards multigap superconductivity in $LaPt_2Si_2$. Moreover, NMR investigation on isostructural $SrPt_2As_2$ shows a Hebel-Slichter coherence peak of $1/T_1$ below T_c [21] which indicates isotropic gap structure in the system. Hence, observation of two gaps in $LaPt_2Si_2$ is unusual among the existing members of $CaBe_2Ge_2$- type structure exhibiting coexistence of CDW and superconductivity. However, recent reports discussing NMR studies [38,55] on $LaPt_2Si_2$ are limited down to 5 K. So, NMR investigations probing the superconducting state in $LaPt_2Si_2$ will be worthwhile.

Now, considering London theory [56] $\lambda_L(0)$ can be related to the effective quasiparticle mass (m^*) and the superfluid density (n_s) by the relation $\lambda_L(0) = m^*c^2/4\pi n_s e^2$ where $m^* = (1+\lambda_{el-ph})m_e$. The value of electron phonon coupling constant λ_{el-ph} which can be derived from McMilan's relation, was already estimated to be 0.53 in our earlier report [17]. Taking this value of λ_{el-ph}, we estimated n_s for different models which are presented in Table I.

ZF-μSR spectra for temperatures above and below T_c are presented in Fig. 4. Here, the muon-spin relaxation, observed in the ZF-μSR spectra, is possibly due to static, randomly oriented local fields associated with the nuclear moments at the muon site. Observed ZF-μSR spectra can be well illustrated using a damped Gaussian Kubo-Toyabe (KT) function, $G_{ZF}(T) = A_1 G_{KT}\exp(-\Lambda t) + A_{BG}$, where A_1 is the initial asymmetry, A_{BG} is the temperature independent background originating from the muons stopping in the sample holder, Λ is the electronic relaxation rate, and G_{KT} is the Gaussian Kubo-Toyabe (KT) function which is expected from an isotropic Gaussian distribution of randomly oriented static (or quasistatic) local fields at the muon sites and is defined as [57], $G_{KT} = [\frac{1}{3} + \frac{2}{3}(1-\sigma_{KT}^2 t^2)\exp(-\frac{\sigma_{KT}^2 t^2}{2})]$, with σ_{KT} being the muon depolarization rate. It is evident from Fig. 4 that ZF-μSR spectra collected above and below T_c show no noticeable change in the relaxation rates. This observation suggests that the time-reversal symmetry is preserved upon entering the superconducting state. On the other hand, small application of a longitudinal magnetic field of just 25 G (see inset of Fig. 4) confiscates any relaxation due to nuclear static fields and is sufficient to fully decouple the muons from this relaxation channel.

IV. CONCLUSION

In summary, we have investigated the superconducting gap structure of a CDW SC $LaPt_2Si_2$ having $T_c = 1.6$ K using TF, ZF, and LF muon spin rotation/relaxation measurements. We have determined the temperature dependence of muon depolarization rate due to the formation of flux line lattice in the superconducting state, by analyzing TF μSR data. Our analysis suggests that the superconducting gap structure in $LaPt_2Si_2$ can be best fitted with two gap s wave model ($s+s$ wave) rather than an isotropic gap s wave model. This conclusion is in agreement with the specific heat analysis which also indicates multigap superconductivity in $LaPt_2Si_2$. On the other hand, ZF data do not show any indication of TRS breaking. Further investigations using other microscopic techniques such as tunnel diode oscillator or scanning tunneling microscopy on good quality single crystals probing the presence of multiple superconducting gaps will be very interesting.

ACKNOWLEDGMENTS

We thank Dr. R. Kulkarni, Prof. S. K. Dhar, Prof. A. Thamizhavel and Mr. K. Panda for cooperation and useful discussions. We also thank Dr. B. Samanataray for help with the analysis of specific heat data. D.D. would like to thank Newton funding for providing travel support to carry out μSR experiments. A.B. would like to acknowledge DST India, for an Inspire Faculty Research Grant (DST/INSPIRE/04/2015/000169). D.T.A. would like to thank CMPC-STFC, Grant No. CMPC-09108, for financial support.

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