Evidence of nodal superconductivity in LaFeSiH

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Evidence of nodal superconductivity in LaFeSiH. By using the complementary techniques of muon spin rotation, tunneling diode oscillator and density functional theory, we unveil the nodal character of the superconducting gap of this novel high-temperature superconductor. Even if the topology of the computed Fermi surface is compatible with the $s_{\pm}$-wave case with accidental nodes, its nesting and orbital-content features may eventually result in a $d$-wave state, more unusual for high-temperature superconductors of this class.

INTRODUCTION

The superconducting energy gap is a hallmark of superconductivity at the level of the electronic structure [1]. Furthermore, the symmetry of the gap function is intimately linked to the microscopic interactions that yield the Cooper pairing, thus providing key information about the mechanism behind superconductivity. Iron-based superconductors have proven to be a distinct class of unconventional superconductors [2] in which the gap symmetry can be tuned by means of external control parameters such as doping, pressure, or disorder [3, 4]. In view of their distinct multiband features, it was soon realized that the so-called $s_{\pm}$-wave gap with a sign change between electron and hole pockets in the Fermi surface is the natural candidate for the gap function in most of these materials [5]. In this case, doping, for example, can lead to enhanced anisotropy by means of various effects such as the modification of intraband Coulomb interactions and changes in the orbital weights on the Fermi surface. At the same time, it was also realized that the $d$-wave pair channel is a strong competitor to the $s_{\pm}$-wave one [6]. In this case the key role is played by the hole pockets where the gap function displays symmetry-imposed nodes. In fact, a strong tendency towards $d$-wave pairing, even dominating over the $s$-wave one, has been found in various models suited for 1111 systems, especially towards the overdoped limit [6–8]. These considerations explain the general trends observed in the Fe-based superconductors, including the controlled changes reported in BaFe$_2$(As,P)$_2$ [9] and (Ba,Rb)Fe$_2$As$_2$ [10].

Here, we investigate the gap structure of the novel superconductor LaFeSiH with $T_c \sim 10$ K in its parent phase [11]. This system is the first silicide in the family of Fe-based superconductors, whose unconventional mechanism of superconductivity is yet to be elucidated [12]. Compared to LaFeAsO, the topology of the Fermi surface is essentially preserved in LaFeSiH at the expense of an increased 3D character that reduces considerably the nesting. To determine the properties of the corresponding superconducting gap we measured the magnetic penetration depth $\lambda$. The temperature dependence of this fundamental quantity maps the excited quasiparticles, and hence the structure of the superconducting gap. Specifically, we performed muon spin rotation and relaxation ($\mu$SR) experiments and used tunnel diode oscillators (TDO) to determine $\lambda$. Details are provided in the Methods section. While the $\mu$SR technique provides a direct access to $\lambda$ by probing the magnetic field distribution in the vortex state [13, 14], the TDO method enables the collection of a large density of points with very high resolution, and hence a very precise determination of the changes in $\lambda$ in the Meissner state [15]. These complementary techniques are supplemented with density-functional-theory (DFT) calculations, from which we compute the zero-temperature penetration depth $\lambda(0)$ in the London approximation and rationalize the nodal behavior observed in our measurements as a function of temperature.

RESULTS

$\mu$SR experiment

Fristly, we report the $\mu$SR experiment. Figs. 1 (a) and (b) show the transverse-field $\mu$SR (TF-$\mu$SR) asymmetry spectra measured in the normal state of LaFeSiH at 20 K and in the superconducting state at 0.3 K. The damping of the asymmetry oscillations observed in the normal state is very small, which indicates that these oscillations are mainly due to nuclear contributions with a distribution of the internal field that is extremely uniform for
the applied field. In the superconducting state, in contrast, the damping is substantially higher, as expected from inhomogeneous field distribution created by the superconducting vortices.

The red lines in Fig. 1 (a) and (b) illustrate the fits of the TF-$\mu$SR data according to Eq. (1). The parameters $\theta$ and $A_{bg}=0.18$ were estimated by fitting to the 20 K and 0.3 K data and their values were kept fixed in the fitting of the other temperature data points. The parameters $A_\kappa$ and $\omega_{bg}$ were allowed to vary, which nevertheless resulted in nearly temperature-independent parameters. The good agreement of the fits validates the model Eq. (1).

The total depolarization rate $\sigma$ as a function of temperature is shown in Fig. 1(c). The clear increase from $\sigma = \sigma_{nm} = 0.508(1)$ with decreasing temperature confirms the emergence of superconductivity in LaFeSiH. This can additionally be concluded from the substantial decrease in the internal field shown in the inset of Fig. 1 (c). The estimated superconducting contribution $\sigma_{sc}$ is shown in Fig. 1 (d). The $T_c$ derived from this data is $\approx 10$ K, which is slightly higher than the onset observed in the DC magnetic-response measurements on the same samples (see also [11]). Since $\mu$SR has much higher sensitivity with respect to the superconducting volume fraction, this suggests that there is a non-negligible distribution of $T_c$’s within the sample.

The zero-temperature penetration depth $\lambda(0)$ can be directly determined from the TF-$\mu$SR parameter $\sigma_{sc}$. The extrapolation of $\sigma_{sc}$ to zero temperature gives $\lambda(0) = 336$ nm, which is similar to that reported in other Fe-based superconductors. This quantity, however, has to be understood as the effective penetration depth $\lambda_{eff}$ which, in powder samples of very anisotropic layered compounds, can be expected to be dominated by the in-plane penetration depth $\lambda_{ab}$. That is, $\lambda_{eff} \approx 3^{1/4}\lambda_{ab}$ [16] so that $\lambda_{ab} \approx 255$ nm. These values can also be compared with the values expected according to the specific band structure of LaFeSiH. The computed values are reported in Table I, which are perfectly compatible with the zero-temperature penetration depth estimated from the $\mu$SR data.

Fig. 1(d) shows the fits of $\sigma_{sc}(T)$ according to Eq. (2) for different models of the superconducting gap (the fitting parameters are summarized in Table II). Among the one-gap models, the best fit is obtained for the model with $d$-wave symmetry of the superconducting gap. In fact, compared to the $s$-wave models, the $d$-wave one reproduces much better the low-temperature behavior [see Fig. 1(d)]. The overall fit improves slightly by con-
which reveals that the skin depth is larger than the size of the superconducting transition with onset $T_c$. The perfect magnetic susceptibility measured in LaFeSiH as a function of temperature for these two configurations. In order to clarify the situation we performed additional TDO measurements. Considering two-gap models, although in this case too the best fit is obtained when the model includes a $d$-wave component. Thus, the TF-$\mu$SR data gained on LaFeSiH, and in particular its low-temperature behavior, suggests a superconducting gap with $d$-wave symmetry. We note, however, that the fits of the TF-$\mu$SR data yield gap values that are systematically below the BCS weak-coupling limit. For the $d$-wave model for example the gap parameter becomes $\Delta_d = 1.41k_BT_c$, while the $s + d$-wave model gives $\Delta_s = 1.30k_BT_c$ and $\Delta_d = 0.57k_BT_c$, both below their corresponding BCS weak-coupling limit. Since these parameters turn out to be unphysical, the TF-$\mu$SR analysis reveals that there is an additional ingredient playing a role.

TDO measurements

In order to clarify the situation we performed additional TDO measurements. In Fig. 2 (a) we show the AC magnetic susceptibility measured in LaFeSiH as a function of the temperature for these two configurations. The perfect diamagnetic behavior observed in both cases confirms the superconducting transition with onset $T_c \approx 10$ K. In the normal state above $T_c$, the TDO signal becomes constant, which reveals that the skin depth is larger than the size of the sample. From this data, the normal-state resistivity is estimated as $> 20 \ \mu\Omega \ cm$ in all the measured samples.

The magnetic susceptibility displays essentially the same behavior as a function of the temperature irrespective of the direction of the applied field. This suggests that the anisotropy between in-plane and out-of-plane superconducting properties is such that $\Delta\lambda_{ab} > \Delta\lambda_{a}/30$, so that the signal is always dominated by $\Delta\lambda_{ab}$ due to the aspect ratio of the samples ($d/w \sim 1/30$). This is in fact in tune with the weak anisotropy obtained in our DFT calculations (see Table I). At the same time, the quantitative agreement between the susceptibility for the two orientations of the magnetic field is rather surprising.

### Table I. Zero-temperature magnetic penetration length of LaFeSiH obtained from DFT calculations in the London approximation. The different rows indicate the values obtained from each Fermi-surface sheet, labelled as in Fig. 3 (sheets 1-3 and 4-5 correspond to the hole-like and electron-like pockets respectively in the $k_z = 0$ plane, i.e. around $\Gamma$ and $M$). $(v^2_f/v) \equiv \frac{1}{S_n} \int dS_n(v^2_f/v)$, where $S_n$ is the corresponding Fermi-surface area.

| FS sheet | $(\frac{\lambda}{n})_1$ [km/s] | $(\frac{\lambda}{n})_2$ [km/s] | $\lambda_a(0)$ [nm] | $\lambda_c(0)$ [nm] |
|----------|-------------------------------|-------------------------------|-------------------|-------------------|
| 1        | 51                            | 12                            | 323               | 698               |
| 2        | 63                            | 5                             | 266               | 951               |
| 3        | 53                            | 66                            | 207               | 186               |
| 4        | 79                            | 18                            | 145               | 305               |
| 5        | 110                           | 10                            | 226               | 759               |

### Table II. Values of the fitting parameters obtained from the $\sigma_{\mu\mu}(T)$ fit for different gap structures. $T_c = 10.0(2) \ K$ was estimated from $s$-wave fit and kept fixed in all other fits.

| Gap model       | $g(\phi)$ | $\Delta_0$(meV) | weight factor | $\chi^2$ |
|-----------------|-----------|-----------------|---------------|---------|
| $d$-wave        | $\cos(2\phi)$ | 1.22            | 1             | 1.20    |
| anisotropic $s$-wave | $\frac{1+\cos(4\phi)}{2}$ | 1.03            | 1             | 3.95    |
| $s$-wave        | 1         | 0.84            | 1             | 4.77    |
| $s + d$-wave    | 1, $\cos(2\phi)$ | 1.09; 0.66    | 0.66; 0.34    | 1.04    |
| $s + s$-wave    | 1         | 1.04; 0.36      | 0.32; 0.68    | 1.10    |

FIG. 2. Magnetic response measured in single-crystal LaFeSiH. (a) AC susceptibility across the superconducting transition ($T_c \approx 10$ K) with the magnetic field applied along the $c$-axis (red) and in the basal $ab$-plane (blue). (b) Change in the magnetic penetration depth $\Delta\lambda$ against $T^2$ in the low-temperature region below $T_c/3$. TDO data is in red for $H \parallel c$ and in blue for $H \perp c$, while the gray circles correspond to the $\mu$SR data. The TDO data has been normalized according to the $\mu$SR data and a vertical offset has been introduced for clarity. Black lines are $T^2$ fits above 2.5 K. The two data sets clearly follow a power-law $T^n$ behavior with $n < 2$ at low temperatures, revealing the presence of line nodes in the superconducting gap.
Also, the drop across the transition is quite broad indicating again a non-negligible distribution of $T_c$’s. These features suggest that the effective geometrical factors are more complex in these samples.

Fig. 2 (b) shows the measured changes in the magnetic penetration depth as a function of $T^2$ in the low-temperature limit. The data clearly follows a power law $T^n$ with an exponent $n < 2$, which is a strong indication of line nodes in the superconducting energy gap [4, 15]. Specifically, when the data is fitted over $T < T_c/3 = 3$ K, the exponent is found $n = 1.8$ for the magnetic field applied along $c$ and 1.7 for the field in the perpendicular direction. These values do not change when the fitting interval is reduced to $T < T_c/6 = 1.5$ K for example, and the same sub-$T^2$ behaviour is observed in other equivalent samples. In fact, the same sub-$T^2$ behavior is also observed in the $\mu$SR data which corresponds to the circles in 2 (b).

The linear-in-$T$ behavior at $T \ll T_c$ of the penetration depth of a superconductor with line nodes is well known to become $T^2$ due to impurity scattering [4, 15]. In the case of a fully gapped superconductor with an unconventional gap structure such as the $s_\pm$ one, the exponential behavior also becomes $T^2$ due to impurity scattering. However, such a possibility is ruled out in our case since we observe an exponent that is always $n \leq 2$. Likewise, an extended $s$-wave with $c$-axis line nodes can be ruled out. The sub-quadratic behaviour, however, is compatible with both a $s_\pm$-wave with accidental nodes (or very deep gap minima) or a $d$-wave with symmetry-imposed nodes.

We note that the measured $\lambda$ does not display any Curie upturn at low temperatures, which indicates that the observed power-law behavior is due to non-magnetic impurities. The degree of disorder that is introduced by these impurities can be quantified by comparing the zero-temperature coherence length $\xi_0 \approx 4.3$ nm [11] to the mean-free path $\ell$. The latter quantity can be estimated from the measured value of the normal-state conductivity and the one computed from DFT as described in the Methods section, which correctly captures the complex multiband features of our system. Thus, $\ell$ is estimated to be $\lesssim 5$ nm. According to this estimate, the samples have to be considered in a borderline case between the clean ($\ell \gg \xi_0$) and the dirty limit ($\ell \ll \xi_0$). This obviously makes the quantitative analysis of the data rather involved, which can explain the above limitations related to the $\mu$SR fits and the TDO geometrical factors. In any case, both these data sets display the same sub-$T^2$ behavior revealing the nodal character of the superconducting gap in LaFeSiH.

**DISCUSSION**

The 1111 compound LaFeAsO provides a reference electronic structure for the Fe-based superconductors. Here, the Fermi surface displays electron and hole pockets that are separated by the wavevector $(\pi, 0)$ (in the 1Fe/unit-cell notation), so that standard considerations on pairing by repulsive interactions suggest a $s_\pm$-wave pair state [5] with a subdominant $d$-wave channel [7, 17]. In this picture the anisotropy in the $s_\pm$ gap function is controlled by several features, notably the 2D vs. 3D character of the Fermi surface and its orbital weights [18, 19]. The strength of the $(\pi, 0)$ interactions, in particular, has a strong dependence on the presence/absence of a $d_{x^2-y^2}$ band near the Fermi level (eventually determined by the actual lattice structure), which then controls the nodeless vs. nodal character of the $s_\pm$ state and can even promote the $d$-wave one [20].

Compared to LaFeAsO, the electronic band structure of LaFeSiH is visibly more 3D as illustrated by the Fermi surface cuts shown in Fig. 3. This is largely due to the prominent dispersion hole pockets — i.e. the inner Fermi-surface sheets 1, 2 and 3 — along the $c$ axis. As a result, the overall nesting between electron and hole pockets is drastically deteriorated, which is highly detrimental for the fully gapped $s_\pm$ pairing and can introduce accidental nodes [20]. In addition, the $d_{x^2-y^2}$ character of the outer sheets is absent, which also goes in the same direction. The $d$-wave channel, in contrast, is mainly linked to the electron pockets (sheets 4 and 5) which better retain its propitious features. These considerations support our
experimental finding of a nodal superconducting gap in LaFeSiH, including the comparatively rare $d$-wave case [6, 21] among the Fe-based superconductors as a possible candidate.

In conclusion, we have determined the magnetic penetration depth of the novel iron-based superconducting silicide LaFeSiH as a function of the temperature in the vortex and in the Meissner state using muon-spin rotation and tunnel-diode oscillators. The observed power-law behavior reveals the presence of low-energy excitations characteristic of nodal superconductivity. The effective zero-temperature value is found to be $\lambda(0) = 336$ nm, in agreement with DFT calculations. The specific features of the electronic band structure of LaFeSiH suggest a prominent role of the electron pockets and accordingly a $d$-wave superconducting gap with accidental nodes (or more generally deep gap minima) is also compatible with our experimental findings. This outlines an analogy to the overdoped deep gap minima) is also compatible with our experimental finding of a nodal superconducting gap in high-temperature superconductors [21]. Furthermore, if $s$- and $d$-wave superconductors, where $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function and $\phi$ is the azimuthal angle across the Fermi surface. Here the superconducting gap function is conveniently expressed as $\Delta(T, \phi) = \Delta_0 \Gamma(T/T_c) g(\phi)$. Thus, its temperature dependence can be taken as $\Gamma(T/T_c) = \tanh(1.82(1.018(T_c/T - 1))^{0.51})$, while its angular dependence can be assumed to be $g(\phi) = 1$ for an isotropic $s$-wave gap, $g(\phi) = \frac{1}{2} (1 + \cos \phi)$ for an anisotropic one, and $g(\phi) = \frac{1}{2} (1 + \cos 2\phi)$ for $d$-wave gap with line nodes for example [14, 24]. This model is readily extended to the multi-gap case.

**METHODS**

Sample preparation

The LaFeSiH powder sample for the TF-$\mu$SR experiment was obtained as described in [11]. From this powder, small single-crystals were singled out for the TDO measurements. The geometry of the selected crystals corresponds to the slab geometry with typical thicknesses $2d \sim 10\mu$m in the $c$ direction and planar dimensions $2w \sim 300\mu$m.

$\mu$SR experiment and analysis

The $\mu$SR experiment was carried out using the MuSR spectrometer at ISIS Facility, UK. Thus, we measured the muon spin depolarization that results from the application of a magnetic field of 30 mT ($> H_{c1}$, see [11]) in the transverse-field configuration (TF-$\mu$SR). The depolarization rate has a component due to the nuclear magnetic contributions of the sample. In addition, if the sample is a type-II superconductor, the depolarization rate is expected to develop an extra contribution due to the inhomogeneous distribution of magnetic field in the vortex state [13], which is directly linked to the magnetic penetration depth $\lambda$. Thus, in the simplest case, the TF-$\mu$SR asymmetry spectrum can be modeled as [13, 14]

$$ A(t) = A_s e^{-\sigma^2 t^2/2} \cos(\omega_s t + \theta) + A_{bg} \cos(\omega_{bg} t + \theta). \quad (1) $$

The first term in this expression describes the oscillations (with relaxation) produced by the sample while the second accounts for the background oscillations (without relaxation) due to e.g. the Ag-sample holder, with $\theta$ is the phase which is related to the detector geometry. In the first term, the total depolarization rate $\sigma$ reads $\sigma = \sqrt{\sigma_{nm}^2 + \sigma_{sc}^2}$ where $\sigma_{nm}$ and $\sigma_{sc}$ represent the aforementioned nuclear and superconducting vortex-lattice contributions respectively. Furthermore, for a triangular vortex-lattice in a type-II superconductor such that $\kappa = \lambda/\xi \gg 70$ and $0.13/\kappa^2 \ll H/H_{c2} \ll 1$, the superconducting part reduces to $\sigma_{sc}^2 = 3.71 \times 10^{-3} \frac{\sqrt{\lambda}}{\lambda}$, where $\gamma_\mu$ is the muon gyromagnetic ratio and $\phi_0$ is the flux quantum [13, 22, 23].

The $\sigma_{sc}$ obtained in this way is therefore directly related to $\lambda$, which we further exploited to determine the superconducting gap $\Delta$ and the pairing symmetry. The temperature dependence of $\sigma_{sc}$, in particular, is such that $\frac{\sigma_{sc}(T)}{\sigma_{sc}(0)} = \frac{\lambda^2(T)}{\lambda^2(0)}$. This ratio is customarily modeled within the London approximation as

$$ \frac{\sigma_{sc}(T)}{\sigma_{sc}(0)} = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_0^\infty \frac{\partial E}{\partial \phi} \sqrt{E^2 - \Delta(T, \phi)^2} \phi dE d\phi, \quad (2) $$

for $s$- and $d$-wave superconductors, where $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function and $\phi$ is the azimuthal angle across the Fermi surface. Here the superconducting gap function is conveniently expressed as $\Delta(T, \phi) = \Delta_0 \Gamma(T/T_c) g(\phi)$. Thus, its temperature dependence can be taken as $\Gamma(T/T_c) = \tan(1.82(1.018(T_c/T - 1))^{0.51})$, while its angular dependence can be assumed to be $g(\phi) = 1$ for an isotropic $s$-wave gap, $g(\phi) = \frac{1}{2} (1 + \cos \phi)$ for an anisotropic one, and $g(\phi) = \frac{1}{2} (1 + \cos 2\phi)$ for $d$-wave gap with line nodes for example [14, 24]. This model is readily extended to the multi-gap case.

**TDO measurements**

We used a high stability LC oscillator with resonant frequency 13 MHz driven by a tunnel diode in a $^3$He refrigerator. Thus, we measured the relative shift of the resonant frequency $\Delta f/\Delta f_0$ which is directly related to the AC magnetic susceptibility $\chi'$ and hence $\Delta \chi = \chi(T) - \chi(0)$ (here $\Delta f_0$ is the frequency shift obtained when the sample is completely extracted from the coil at the base temperature, while the factor of proportionality is defined by the TDO effective dimension of the sample) [15, 24–26].

The geometry of the measured crystals corresponds to the slab geometry with typical thicknesses $2d \sim 10\mu$m in the $c$ direction and planar dimensions $2w \sim 300\mu$m. Thus, the TDO effective sample dimension is expected to be $\sim 0.2w$ when the magnetic field is applied along the $c$ axis and $\sim d$ when it is perpendicular to $c$ [27]. Furthermore, if $H \parallel c$ then the screening supercurrents flow entirely in the $ab$-plane and hence the in-plane penetration depth $\lambda_{ab}$ is probed. However, if $H \perp c$ the screening is due to supercurrents flowing both in-plane and out-of-plane so that the mixture $\lambda_{ab} + \frac{d}{w} \lambda_c$ containing the contribution due to the out-of-plane penetration depth $\lambda_c$ is probed.

**DFT calculations**
We performed DFT calculations using the FLAPW method as implemented in the WIEN2K package [28] with the PBE exchange-correlation functional [29]. Specifically, we considered the low-temperature structure reported of LaFeSiH in [11], with muffin-tin radii of 2.30, 2.10, 2.20 and 1.20 a.u for La, Fe, Si and H atoms respectively, and a plane-wave cutoff $R_{\text{MT}}K_{\text{max}} = 5.0$ in our spinless calculations. The integration over the Brillouin zone was performed using a $15 \times 15 \times 7$-k mesh, while the Fermi velocity was computed using a denser $64 \times 64 \times 32$ k-mesh as $v = p/m_e$ with $p$ being the expectation value of the momentum operator and $m_e$ the electron mass.

From these calculations we further computed the penetration depth in the London approximation according to the formula $(\lambda^2)_{ij}^{-1}(0) = \mu_{\text{e}} e^2 \pi \int_{\text{FS}} dS_{\nu} / \sqrt{v_F}$ (see e.g. [15, 27]). Here $\nu$ is the Fermi velocity and the integral is over the Fermi surface. In these calculations In addition, we also computed the conductivity integral is over the Fermi surface. In these calculations ($e^2/\Omega_0) \int_{BZ} v_i(k) v_j(k) \delta(\varepsilon(k) - \varepsilon_F) dk$ within the Boltzmann transport theory [30]. Here $\Omega_0$ is the volume of the first Brillouin zone and the relaxation time $\tau$ gives the mean-free path as $\ell = v_F \tau$.

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