Quasi-two-dimensional Fermi surfaces of the heavy-fermion superconductor Ce2PdIn8

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We report low-temperature de Haas–van Alphen (dHvA) effect measurements in magnetic fields up to 35 T of the heavy-fermion superconductor Ce_2PdIn_8 . The comparison of the experimental results with band-structure calculations implies that the 4*f* electrons are itinerant rather than localized. The cyclotron masses estimated at high field are only moderately enhanced, $8m_0$ and $14m_0$, but are substantially larger than the corresponding band masses. The observed angular dependence of the dHvA frequencies suggests quasi-two-dimensional Fermi surfaces in agreement with band-structure calculations. However, the deviation from ideal two-dimensionality is larger than in CeCoIn₅, to which $Ce₂PdIn₈$ bears a lot of similarities. This subtle distinction accounts for the different superconducting critical temperatures of the two compounds.

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I. INTRODUCTION

The appearance of unconventional superconductivity in the vicinity of a quantum critical point (QCP) is a common trend in Ce-based heavy-fermion (HF) compounds. A more recent and still somewhat controversial issue is the influence of the Fermi-surface (FS) dimensionality on unconventional superconductivity. Indeed, reduced dimensionality of the FS leads to nesting-type magnetic instabilities [\[1\]](#page-4-0) and thus enhances the superconductivity $[2,3]$. The exact knowledge of the FS topology of HF systems is, therefore, essential. In addition, this information allows distinguishing if the *f* electrons are itinerant or localized, i.e., whether they contribute to the FS or not.

 $Ce₂PdIn₈$ is a recently discovered HF superconductor with $T_c = 0.7$ K and a nonmagnetic ground state [\[4,5\]](#page-4-0). Non-Fermi-liquid behavior was observed in both macroscopic [\[6–10\]](#page-4-0) and microscopic [\[11,12\]](#page-4-0) measurements at low temperature, implying that $Ce₂PdIn₈$ is located very close to a QCP. It was further suggested that a two-dimensional (2D) spin-densitywave-type QCP is induced by magnetic field near the upper critical field, $H_{c2} \approx 2 \text{ T} [9]$ $H_{c2} \approx 2 \text{ T} [9]$. Unconventional superconductivity was demonstrated to be due to antiferromagnetic quantum fluctuations [\[13\]](#page-4-0). These unusual properties are strikingly similar to those of the well-studied HF superconductor $CeCoIn₅$ [\[14–17\]](#page-4-0), which is also located very close to a QCP at ambient pressure. However, the superconducting critical temperature $T_c = 2.3 \text{ K}$ of CeCoIn₅ [\[18\]](#page-4-0) is considerably higher than that of Ce₂PdIn₈.

 $Ce₂PdIn₈$ crystallizes into a tetragonal $Ho₂CoGa₈$ -type crystal structure with space group *P*4*/mmm*. It belongs to the larger family of $Ce_nTIn_{3n+2}(T)$: transition metal, $n = 1, 2$, and ∞) systems, containing a sequence of *n* CeIn₃ layers interca-

In this paper, we report high-field de Haas–van Alphen (dHvA) measurements of $Ce₂PdIn₈$. The observed FS is quasi-2D; however, we find that the more three-dimensional crystal structure of $Ce₂PdIn₈$ relative to $CeCoIn₅$ leads to a reduced two-dimensionality of the FS topology. We argue that this can explain the difference in the superconducting critical temperatures of the two compounds.

II. EXPERIMENTAL DETAILS

Single crystals were grown by the self-flux method [\[4\]](#page-4-0), and we have confirmed by specific-heat measurements that they are not contaminated by CeIn₃. The dHvA measurements were performed using a torque cantilever magnetometer mounted in a top-loading dilution refrigerator equipped with a low-temperature rotator. Magnetic fields, *B*, up to 35 T generated by LNCMI-Grenoble resistive magnets were applied at different angles between the [001] and [100] directions.

III. RESULTS AND DISCUSSION

Figure [1](#page-1-0) shows the oscillatory torque after subtracting a nonoscillating background and the corresponding Fourier transform in $Ce₂PdIn₈$. Four fundamental frequencies, denoted *ζ* , *η*, *α*1, and *α*2, are observed when *B* is applied close to the

lated by a $T\text{In}_2$ layer along the *c* axis. While cubic CeIn₃ (*n* = ∞) is a completely isotropic system, the layered structures with $n = 1$ and 2 are expected to lead to anisotropic properties and quasi-2D FSs. Indeed, quasi-2D FS sheets were observed in both $n = 1$ systems CeCoIn₅ [\[19,20\]](#page-4-0), CeIrIn₅ [\[21\]](#page-4-0), and CeRhIn₅ [\[22,23\]](#page-4-0), and the $n = 2$ compound Ce₂RhIn₈ [\[24,25\]](#page-4-0). The degree of two-dimensionality is expected to be larger in monolayer systems with alternating layers of CeIn₃ and $TIn₂$ than in their bilayer counterparts, in which two $Cefn₃$ layers are separated by one $T\ln_2$ layer.

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FIG. 1. Fourier spectrum of the high-field dHvA oscillations (inset) in $Ce₂PdIn₈$ for magnetic field applied at $4°$ off the *c* axis at 30 mK.

c axis. The oscillations were traced up to 60◦, where their amplitude decreased below the noise level.

To figure out whether the *f* electrons are itinerant or localized in $Ce₂PdIn₈$, we performed band-structure calculations for both Ce_2PdIn_8 and La_2PdIn_8 , the latter corresponding to the Ce compound with localized *f* electrons. For both compounds, the calculations were carried out using a full potential augmented plane wave method with the local density approximation (LDA) for the exchange-correlation potential. As crystals of La_2PdIn_8 are currently unavailable, the lattice parameters of $Ce₂PdIn₈$ were used for the $La₂PdIn₈$ calculations. The resulting FSs are shown in Fig. 2.

Given the layered crystal structure, it is not surprising that some of the calculated FS sheets are quasi-2D in both $Ce₂PdIn₈$ and $La₂PdIn₈$. The details of the FSs are, however, clearly different. In contrast, the topology of the 4*f* -itinerant FS of CeCoIn₅ is similar to the $4f$ -localized FS of LaRhIn₅ and CeRhIn₅ [\[22,23\]](#page-4-0), where the two FSs differ mainly by

size. It should be noted that $CeCoIn₅$ is a compensated metal with equal carrier numbers of electrons and holes, while $LaColn₅$ is an uncompensated metal. On the other hand, both $Ce₂PdIn₈$ and $La₂PdIn₈$ are compensated metals. As seen in Fig. 2, the charge-carrier number given by the FS volume in $Ce₂PdIn₈$ is about two times smaller than that in $La₂PdIn₈$.

Figure $3(a)$ shows the experimentally observed angular dependence of the dHvA frequencies in $Ce₂PdIn₈$ together with the results of band-structure calculations based on the 4*f* itinerant band model. Experimental and calculated frequencies and effective masses are also shown in Table [I.](#page-2-0) The agreement between the experimentally observed *α* branches and those of the calculations is excellent. Not only are the angular dependencies the same, but even the absolute values agree very well. This implies that both the topology and the size of the calculated FS sheet reproduce the experimental results exceptionally well. The α branches correspond to the quasi-2D FS of band 73, as shown in Fig. 2. Regarding the measured lower (*<*2 kT) dHvA frequencies, there is also a very good agreement between the experimental and calculated branches. These branches originate mostly from rather isotropic parts of the FS in band 72. The calculated *β* branches originating from complicated sheets in band 72 were, however, not observed in the experiment. This is probably caused by a strongly enhanced effective mass or an unfavorable curvature factor for detecting the dHvA signal. For comparison, in Fig. $3(b)$ we plot the experimental results obtained in $Ce₂PdIn₈$ together with band-structure calculations for La_2PdIn_8 . In this case, the two are obviously at odds with each other. In particular, only one quasi-2D FS was observed in the experiment, while the calculations predict two of them for La_2PdIn_8 , which should be easy to detect.

The comparison of the experimentally observed dHvA frequencies with the results of the LDA band-structure calculations thus gives clear evidence for a quasi-2D FS with

FIG. 2. Calculated FSs of Ce₂PdIn₈ (left). Calculated FSs of La₂PdIn₈ (right) are also shown for comparison.

FIG. 3. (Color online) Angular dependence of the experimentally observed dHvA frequencies in Ce₂PdIn₈ (open circles) is shown together with the results of band-structure calculations (solid circles) performed for (a) Ce_2PdIn_8 and (b) La_2PdIn_8 . The latter correspond to Ce_2PdIn_8 with localized 4f electrons and are shown for comparison. Very low calculated dHvA frequencies that correspond to small FS pockets are not shown for clarity.

itinerant f electrons in $Ce₂PdIn₈$. The same conclusion was drawn for CeCoIn₅ $[19,20]$.

We, alternatively, calculated the band structure using the local spin-density approximation with the relativistic version of the full-potential local orbital method $[26]$ for Ce₂PdIn₈. This also suggests quasi-2D FS, but the agreement with the experimental results is not as good.

The effective masses shown in Table I were determined by fitting the temperature dependence of the oscillatory amplitude by the standard Lifshitz-Kosevich formula [\[27\]](#page-4-0). This was done for the magnetic field applied at 4◦ off the *c* axis. Due to the small amplitudes of the oscillations the field range

TABLE I. Experimental and calculated dHvA frequencies and effective masses in $Ce₂PdIn₈$ for magnetic field along the *c* axis.

Branch	Experiment		Calculation	
	F (kT)	$m^*/m_0^{\rm a}$	F (kT)	m_b/m_0
γ			0.34	1.55
δ			0.43	0.42
ζ	0.78	8.4 ± 0.4	0.69	1.24
η	1.2		1.07	2.27
α_2	3.26		2.82	0.81
α_1	4.82	14 ± 1	4.82	2.12
β_2			7.27	2.81
β_1			13.08	5.2

^aThe effective masses were measured with magnetic field applied at 4◦ off the *c* axis.

from 28 to 34.5 T was used for the analysis. Even for such high fields, the effective masses of only two branches, *ζ* and α_1 , could be reliably determined. The obtained values are $(8.4 \pm 0.4)m_0$ and $(14 \pm 1)m_0$, respectively. The effective mass of the α_1 branch corresponding to the quasi-2D sheet of the FS is comparable to the values, $8m_0-18m_0$, reported for the quasi-2D FS of CeCoIn₅ $[19,20,28]$. This implies a similar degree of hybridization between the *f* and conduction electrons. The detected effective masses are, however, by far too small to account for the huge value of the electronic specific heat coefficient, γ , of the order of 1 J/K² mol just above the superconducting transition $[4,5,9]$. Presumably, the effective masses of the β branches, which are not observed here, are strongly enhanced. Indeed, already the calculated band masses of the *β* branches are higher than those of the other branches (see Table I). On the other hand, the Sommerfeld coefficient of $Ce₂PdIn₈$ is similar to that of $CeCoIn₅$ [\[18\]](#page-4-0), where the effective masses were reported to strongly decrease with magnetic field $[19]$. While the observed dHvA oscillations in Ce₂PdIn₈ are not strong enough to perform the field-dependent analysis of the effective masses, they can also be expected to decrease with magnetic field. This assumption is supported by the experimentally observed field dependence of the T^2 coefficient in the resistivity [\[6\]](#page-4-0) and of the Sommerfeld coefficient of the specific heat [\[9\]](#page-4-0) above the upper critical field.

As shown in Fig. [4,](#page-3-0) the major FS sheets of both $Ce₂PdIn₈$ and CeCoIn₅ are quasi-2D corrugated cylinders extending along the [001] direction. As many of the physical properties of HF materials strongly depend on the FS dimensionality, the key question here is which FS is more 2D, i.e., which

FIG. 4. Comparison of the calculated quasi-2D FSs of CeCoIn₅ and Ce₂PdIn₈.

amplitude of the corrugation is smaller. For both $Ce₂PdIn₈$ and CeCoIn5, the FSs experimentally determined through dHvA measurements are in excellent agreement with calculated ones. This is, however, not always the case. That is why we introduce a quantitative criterion of a quasi-2D FS deviation from an ideal cylinder: $\Delta = (S_{\text{max}} - S_{\text{min}})/S$, where S_{max} and S_{min} are the maximum and minimum extremal cross sections, respectively, and *S* is the average cross section of the warped cylinder. For an ideal cylinder, $\Delta = 0$. Since the extremal cross sections, S_i , of the FS are proportional to the dHvA frequencies F_i , *S* can be replaced by *F* measured with field along [001] to determine Δ experimentally. In Ce₂PdIn₈, with the two frequencies, α_1 and α_2 , listed in Table [I,](#page-2-0) this yields $\Delta = 0.386$. The dHvA effect measurements in CeCoIn₅ revealed three extremal cross sections of the quasi-2D FS [\[19,20,28\]](#page-4-0). The reported values of the dHvA frequencies are slightly different, yielding the average value of $\Delta = 0.221$. This implies that the deviation from the ideal 2D FS is much smaller in CeCoIn₅ than in $Ce₂PdIn₈$. This is expected as well from the more three-dimensional crystal structure of $Ce₂PdIn₈$ as compared to CeCoIn₅.

The corresponding larger anisotropy of $CeCoIn₅$ as compared to $Ce₂PdIn₈$ accounts for the higher superconducting critical temperature of $CeCoIn₅$. In fact, 2.3 K in $CeCoIn₅$ is the highest T_c among all the known Ce-based HF materials. Remarkably, the FS of $CeCoIn₅$ is the most 2D-like as compared to its Ir and Rh analogs $[20]$. The FS of CeRhIn₅, however, changes at its critical pressure $P_c \simeq 2.4 \text{ GPa}$ [\[29\]](#page-4-0),

and the reported dHvA frequencies yield $\Delta = 0.17$ above P_c . This value is similar to that of CeCoIn₅ at ambient pressure. In CeRhIn₅, superconductivity emerges around P_c , where $T_c =$ 2.1 K [\[30\]](#page-4-0), a value close to that of CeCoIn₅. Regarding CeIrIn₅, experimentally observed dHvA frequencies [\[21\]](#page-4-0) result in $\Delta =$ 0.269, a value in between those for $CeCoIn₅$ and $Ce₂PdIn₈$. However, $T_c = 0.4$ K of CeIrIn₅ [\[31\]](#page-4-0) cannot be compared directly to the critical temperatures of $CeCoIn₅$ and $Ce₂PdIn₈$, as CeIrIn₅ is located farther away from a QCP $[32]$. When CeIrIn₅ is tuned to a QCP by Rh substitution, T_c increases to about 1 K [\[33\]](#page-4-0) and is likely to be reduced due to disorder as compared to pure compounds. Consistently with Δ , this value also falls in between those for $CeCoIn₅$ and $Ce₂PdIn₈$. Unfortunately, there is currently no information about the FS of Rh-substituted CeIrIn₅. While the FS dimensionality is not the only factor that determines T_c in HF superconductors, it is certainly a significant one. Indeed, $T_c = 18.5$ K was reported for PuCoGa₅ [\[34\]](#page-4-0), which is the highest among those yet observed in *f* -electron materials. Remarkably, the calculated FS of PuCoGa₅ consists of three corrugated cylinders $[35]$, although the degree of corrugation is relatively high with Δ being 0.448, 0.359, and 0.66, respectively. However, the results of these calculations are still to be confirmed experimentally.

IV. SUMMARY

In summary, our high-field dHvA investigation of $Ce₂PdIn₈$ combined with band-structure calculations evidence the existence of a quasi-2D FS with itinerant *f* electrons in this compound. The comparison of the FS topology of $Ce₂PdIn₈$ and $CeCoIn₅$ implies that the FS of the latter compound is much closer to an ideal cylinder characteristic for a 2D case. The difference in the FS dimensionality accounts for different superconducting critical temperatures of the two compounds, which are both located in close vicinity to a QCP and have a similar degree of hybridization between the 4*f* and conduction electrons. It would be interesting to apply the quantitative criterion of the FS two-dimensionality we introduced here to other HF materials with quasi-2D FSs. In particular, the criterion can be used to verify the theoretical prediction about the influence of the FS dimensionality on the type of quantum criticality in HF compounds [\[36–39\]](#page-4-0). Another interesting question is whether magnetic fields themselves affect the FS dimensionality in $Ce₂PdIn₈$ in particular and other quasi-2D HF materials in general. Zero-field angle-resolved photoemission spectroscopy measurements in $Ce₂PdIn₈$ would be very useful to address this issue.

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