Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B Magnetic phase diagram and superconducting parameters

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Abstract. Using an ac susceptometry technique, $H-T$ phase diagrams were determined for various Pt contents in the noncentrosymmetric superconducting system Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B, from which several superconducting parameters could be extracted. Despite the end compositions having very different band and gap structure, and despite a change from singlet- to triplet-dominant pairing, these superconducting parameters change continuously with cation substitution. Weak-coupling BCS and Ginzburg-Landau relations appear to be obeyed, suggesting that this may be a particularly uncomplicated system for studying the effect of a lack of inversion symmetry.

Most superconductors’ crystal structures contain at least one inversion centre — transposing every atomic position through that point returns the original structure. If an inversion centre is present, the parity of the pairing state’s orbital component dictates the spin component — keeping the electrons’ wavefunctions antisymmetric requires singlet pairing for odd parity and triplet for even parity. This underpins much of our understanding of superconductivity. In noncentrosymmetric superconductors, parity is not a meaningful quantity, singlet and triplet are not eigenstates, and extremely unconventional behaviour is possible. Exotic physics was first found in CePt$_3$Si [1], which has a very high, non-Pauli-limited upper critical field $H_{c2}$, a key signature of novel behaviour in such materials. A number of other noncentrosymmetric superconductors have since been found, including UIr [2], Mo$_3$Al$_2$C [3], B-doped SiC [4], and 1-1-3 silicides like CeRhSi$_3$ [5], BaPtSi$_3$ [6], and CaIrSi$_3$ [7, 8].

Without inversion, otherwise-forbidden spin-orbit terms can split the band structure and Fermi surfaces by spin orientation, leading to a spin imbalance at the Fermi surface and, for large splittings, to pairing largely within each Fermi-surface sheet [9–11]. The most exotic behaviour occurs in external fields [12–16]. Perhaps because significant spin-orbit splitting is required, exotic physics has been seen mainly in heavy-Fermion lanthanide and actinide compounds, and many theoretical predictions remain unverified.

Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B seems unique in showing unconventional behaviour without heavy Fermions — it appears fully gapped and singlet-dominated for $x = 0$ but exhibits line nodes and strong

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triplet character for \( x = 1 \) \([17, 18]\), with hints of unconventional behaviour from \( x = 0.2 \) \([19]\). Characterizing these fundamental changes may help illuminate the origins of and requirements for exotic noncentrosymmetric-derived behaviour. This paper presents the substitution- and temperature-dependence of parameters that can be extracted from the magnetic phase diagram, looking for deviations from simple, BCS weak-coupling behaviour.

Polycrystalline ingots of \( \text{Li}_2(\text{Pd}_{1-x}\text{Pt}_x)_3\text{B} \) were prepared by arc melting as described elsewhere \([20]\). Pieces of the ingot were measured by \( ac \) magnetic susceptibility by a phase-sensitive mutual-inductance technique in a homebuilt first-derivative coil, mounted in a \(^3\)He refrigerator (Oxford Instruments) in a 9 T magnet. An \( ac \) drive field, parallel to the \( dc \) field, of \( \sim 800 \) Hz and 100 \( \mu \)T rms was used. \( H_{c2} \) was defined as where the diamagnetic signal \( \chi'_{ac} \) reached 5% of its full, zero-field, low-temperature value; example \( \chi'_{ac} \) data for \( x = 0.50 \) are shown in Fig. 1a, and the \( H–T \) phase diagram extracted from these data is shown in Fig. 1b.

\[ H_{c2}(0) \text{ and } T_c \text{ are shown as a function of substitution in panel c) (as also shown in [21]), with fits as described in the text.} \]

From the extrapolated \( H_{c2}(0) \)s, shown in Fig. 1a, as a function of cation substitution along with \( T_c \), the relation \( \mu_0 H_{c2}(0) = \Phi_0/2\pi \xi_0^2 \) (\( \Phi_0 \) being the flux quantum) for orbital-limiting fields may be used to calculate a zero-temperature coherence length \( \xi_0 \), assuming orbital-limiting effects to be dominant. These values, shown in Fig. 2a, appear to be linear in cation substitution. This suggests the functional forms \( H_{c2}(0, x) \propto (x + a)^{-2} \) and \( T_c \propto (x + a)^{-1} + \text{Const} \), assuming that the BCS \( \xi_0(x) \propto h v_F/k_B T_c(x) \) relation holds and that the Fermi velocity \( v_F(x) \) is approximately constant. The dashed curves in Fig. 1c show the result of such a fit, with the \( a \) value of \( \approx 1.5 \) taken from the linear fit to \( \xi_0(x) \). That such a simple substitution dependence is observed in a quantity incorporating such parameters as the density of states and the gap (and that the BCS approximations required seem to produce reasonable results) is remarkable in such an unconventional system, and indicates a strikingly smooth transition between two states with very different gap and electronic structure.

\[ \text{Figure 1. a) Example } \chi'_{ac} \text{ data, for } x = 0.50, \text{ and b) the extracted phase diagram (WHH in pink). Data and phase diagrams at other } x \text{ look similar [21].} \]
Assuming the orbital-limiting Ginzburg-Landau form $\mu_0 H_{c2}(T) = \Phi_0 / 2\pi \xi^2(T)$, it is further possible to extract $\xi(T)$ for each $x$ — see Fig. 2b. Due to concerns that the upward curvature at very low fields is an artifact of grain boundary transport, the $T_c$ used for this graph’s horizontal axis was extrapolated from the linear region of each $H$–$T$ phase diagram. The unusual behaviour at the left of Fig. 2b is due to this curvature. The central part of each trace is well-described by a straight line with the slope of $1/T_c^2$ expected from Ginzburg-Landau theory near $T_c$, reinforcing the identification of the upturn as an artifact. In fact, the behaviour remains linear over a wider temperature range than would be expected if WHH dirty-limit predictions (Fig. 1b) were processed the same way for comparison. The curvature could arise from the Pauli depairing, band structure changes [22], strong coupling effects [23], from the $\chi_{ac}$ technique detecting a vortex melting transition rather than $H_{c2}$, or from multiband superconductivity as in MgB$_2$. Indeed, the Fermi surface consists of a large number of distinct sheets, both electron- and hole-like [24], with many opportunities for nesting [23]. In any case, the degree of agreement with simple BCS and Ginzburg-Landau predictions is surprising.

The clear, abrupt changes one might expect between singlet- and triplet-dominated pairing do not appear to be present in this system, in either the $H$–$T$ phase diagrams, $T_c$, $H_{c2}$, or $\xi$. The superconducting parameters evolve smoothly with cation substitution, obeying simple BCS and Ginzburg-Landau expectations, with a coherence length linear in Pt content. Since the transition between the two endmembers appears continuous and gradual, the entire substitution range will be of interest. This also suggests that the apparent onset of novel physics at any particular Pt content may be a coincidence or a characteristic of the probe used. The degree of agreement with simple, well-understood theory will make this system an ideal testbed for predictions of novel noncentrosymmetric physics.

**Figure 2.** Extracted coherence length a) $\xi_0$ [21] and b) $\xi(T)$, assuming the dominance of orbital depairing and using a $T_c$ value extrapolated from the linear portion of each phase diagram. The WHH curve was processed the same way for comparison.
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