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Specific-heat studies of the spin-orbit interaction in noncentrosymmetric $Li_2(Pd_{1-x}Pt_x)_3B$ $(x=0,0.5,1)$ superconductors

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The temperature- and field-dependent specific heat $C(T,H)$ measurement on the noncentrosymmetric superconductors Li_2Pd_3B, Li_2Pt_3B, and $Li_2(Pd_{0.5}Pt_{0.5})_3B$ were carried out over a wide range of T and H. $C(T,H)$ of the limit compounds is found to be distinctly different: within the superconducting phase, the electronic specific heat of the Pd-based compound follows $C_e^S(T<T_c=6.95\text{ K},0\text{ T})=3\exp[-16.4/T]$ J/mol K while for the Pt-based compound $C_e^S(T<T_c=2.56\text{ K},0\text{ T})=\alpha T^2$ mJ/mol K, where $\alpha=7.3$ mJ/mol K^3. An increase in H tends to reduce weakly the exponential argument and the coefficient of the quadratic term. The above-mentioned findings are in agreement with the description that, due to the difference in the strength of the spin-orbit coupling, the pairing symmetry in the Pd- (Pt-) based compound is predominately a spin-singlet (spin-triplet) state. Within the very low-temperature range, the field-induced normal-state of the Pd-based limit manifests $C^N(T,7T>H_{c2})=9.3T+1.1T^3$ mJ/mol K while that of the Pt-based isomorph shows $C^N(T,7T>H_{c2})=9.3T+0.92T^3$ mJ/mol K. For the intermediate alloy $Li_2(Pt_{0.5}Pd_{0.5})_3B$, $C(T,H)$ mimics that of the Pt-based compound; specifically, $C_e^S(T<T_c=3.9\text{ K},0\text{ T})=\alpha T^2$ mJ/mol K where $\alpha=4.6(2)$ mJ/mol K^3. The ratio of the coefficients of the quadratic terms in $Li_2(Pt_{0.5}Pd_{0.5})_3B$ and Li_2Pt_3B, $\alpha_{Pt_{0.5}Pd_{0.5}}/\alpha_{Pt}$, is equal to $Z^2_{Pt_{0.5}Pd_{0.5}}/Z^2_{Pt}$, confirming that the line nodes are driven by the spin-orbit interaction. The finding that these nodes can survive even in a 50% Pd-diluted material gives extra support to the reported claim that the x-dependent evolution of T_c, H_{c1}, H_{c2}, and $(\partial H_{c2}/\partial T)_{T_c}$ is almost linear.

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I. INTRODUCTION

The intermetallic superconductors Li_2Pd_3B and Li_2Pt_3B (Refs. 1–3) crystallize[4] as a cubic structure with space group $P4_332$ wherein the symmetry group does not include the inversion symmetry operation; in particular, the Pt and Pd atoms are situated at noncentrosymmeteric sites. Moreover, the higher atomic numbers of Pt (Z_{Pt}) and Pd (Z_{Pd}) together with the fact that their ratio $(Z_{Pt}/Z_{Pd})^2\approx 3$ emphasize the importance of antisymmetric spin-orbit coupling (ASOC) which, as evident, would be much stronger for the Pt-based compound than its Pd-based isomorph. In general, the absence of inversion symmetry together with a strong ASOC in intermetallic matrices such as $CePt_3Si$,[5] UIr,[6] are shown to give rise to unconventional superconductivity. In contrast to these (magnetic) superconductors, the titled compounds do not show strong electronic or magnetic correlations;[7] as such they are attractive candidates for studying possible exotic superconducting features.[8,9] Indeed, the presence of a strong ASOC in these Li-rich borides is shown to give rise to an admixture of spin-singlet and spin-triplet pairing states.[10,11] That Li_2Pt_3B has a stronger ASOC is manifested as having a predominantly spin-triplet pairing state and, due to the presence of line nodes, a power-law temperature-dependence in its NMR relaxation time[10] $(1/T_1\propto T^3)$ and in its penetration depth[11] $(\Delta\lambda\propto T)$. On the other hand, for Li_2Pd_3B, the thermal evolutions of its specific heat,[12] penetration depth,[11,13] and $1/T_1$ (with a Hebel-Slichter peak)[10] suggest a dominant, weakly anisotropic but fully gapped spin-singlet state.

Considering that in the solid solution $Li_2(Pd_{1-x}Pt_x)_3B$, the Pt and Pd atoms enter substitutionally (and in a random fashion) and that the absence of centrosymmetric operation is maintained across the series, then an increase in x would be accompanied by an increase in the intensity of ASOC, presumably proportional to $[xZ_{Pt}+(1-x)Z_{Pd}]^2$. This, in turn, would enhance the splitting of the degenerate electronic band into two spin-dependent parts[9] leading to an x-dependent degradation of the superconductivity of $Li_2(Pd_{1-x}Pt_x)_3B$ (recall that doping Li_2Pt_3B with Pd leads to an increase in T_c, a feature indicative of a reduction in an alloying-induced depairing effect). Along this line of arguments, the almost linear x-dependent variations[2,3] of T_c, H_{c1}, H_{c2}, and $(\partial H_{c2}/\partial T)_{T_c}$ are being interpreted.[11] To qualify such an interpretation, one should address the following objection: alloying with Pt (the electronic configuration of which differs substantially from that of Pd) would introduce a strong scattering channel which, in addition to those channels inherent in (poly)crystals synthesized by the arc-melting method, would lead to a shorter electron collision time τ and as such the triplet component of the order parameter should be strongly suppressed. If this scenario is correct, then the resulting shorter τ would be reflected as, say, an expressive increase in the temperature-independent, low-temperature resistivity of the solid-solution. The extensive studies of Badica *et al.*[3] does

TABLE I. Some normal and superconducting parameters of Li_2Pd_3B and Li_2Pt_3B. All notations have their usual definitions. Note that while the superconducting parameters of Li_2Pd_3B are substantially enhanced as compared to those of Li_2Pt_3B, their normal-state parameters are almost equal. The discrepancy among these parameters (in particular T_c, γ, and β) and those of the present work is discussed in the text and Fig. 1.

	Experiment parameters (Ref. 12)				Electrodynamic parameters (Refs. 3, 10, and 11)						
Li_2T_3B	T_c (K)	γ (mJ/mol K^2)	θ_D (K)	$\Delta C/\gamma T_c$	T_c (K)	ρ ($\mu\Omega$ cm)	ξ (nm)	l (nm)	$\lambda_0[0\text{ K}]$ (nm)	$2\Delta_0$ (K)	$N(E_F)$ (1/eV/fu)
Li_2Pd_3B	7.5(3)	9.0(1)	221(1)	2.0(3)	6.7	20	9.5	24	190	29.6(1.0)	2.24
Li_2Pt_3B	2.17(5)	7.0(1)	228(1)	1.39(3)	2.43	28	14.5	42	364	7.7(2)	2.9

not support this scenario; rather it showed that the ratio of the normal-state resistivities of $Li_2(Pd_{1-x}Pt_x)_3B$ is almost the same ($\rho_{300}/\rho_{10} \approx 1.4$) for all solid solutions inclusive of the end members (see Fig. 4 of Ref. 3). It is worth adding that the density of state $N(E_F)$ is reported to be 2.9 state/eV formula unit for Pt-based compound while 2.2 state/eV formula unit for Pd-based analog.[17,18] Moreover, the mean free path is calculated[11] to be 240 Å for Li_2Pd_3B and 420 Å for Li_2Pt_3B. Considering a typical Fermi velocity of 3×10^5 m/s [Ref. 17], then the relaxation time is expected to be $\sim 100 \times 10^{-15}$ s.

It is evident from the earlier paragraph that, due to the action of the ASOC, there is a distinct difference between the superconductivity of the otherwise very much similar parent compounds. To appreciate such a distinction, Table I shows some of the reported experimental parameters. Evidently neither γ [as a measure of $N(E_F)$] nor the Debye temperatures could be useful in predicting the differences among the superconductivity of the limit compounds. As the properties of these compounds do not suggest any conventional pair-breaking mechanism, then the above-mentioned variation in the perturbing ASOC would be a reasonable explanation for the strong x-dependency of T_c, H_{c1}, H_{c2}, and $(\partial H_{c2}/\partial T)_{T_c}$.[2,3] Though it is implicitly assumed, power-law relations (or the admixture of spin-singlet and spin-triplet states) have never been verified in any one of these alloyed superconductors. Even for the *limit* compounds, the verification of the power-law relations was carried only for $1/T_1$ and $\Delta\lambda$. Since these power laws form a simple and a handy criterion for the evaluation of the ASOC in these solid-solutions, then it is of interest to verify experimentally the existence or absence of these power laws when these noncentrosymmetric superconductors are subjected to a variation in x. Similarly, the functional dependence of these quantities when H is varied would be a welcome contribution. Such x- and H-dependent results are crucial for the validity of the above-mentioned interpretation of the x-dependence of T_c, H_{c1}, H_{c2}, and $(\partial H_{c2}/\partial T)_{T_c}$. This work reports on the concentration-, thermal-, and field-dependent superconducting specific heat, $C_e^S(T,x,H)$, of $Li_2(Pd_{1-x}Pt_x)_3B$ (x=0.0, 0.5, and 1.0). Particular attention is paid to the existence or absence of the power-law relation[8] $C_e^S(T,x,H) = \alpha(x,H)T^2$. In addition to confirming the distinct difference between the superconductivity of the limit compounds, we observed that even when the strength of the ASOC is varied by 50% Pd dilution, the quadratic thermal evolution is maintained and with a coefficient which scales with the effective atomic number.

II. EXPERIMENT

Samples were prepared by a two-stage arc-melting process (for details, see Ref. 1). Based on our metallurgical, structural, and physical characterizations, these samples are considered to be high-quality single phases. Resistivity and magnetization results (not shown) are in accord with those results given in Refs. 1–3: in particular, the temperature-independent residual resistivity of Li_2Pd_3B, $Li_2(Pd_{1/2}Pt_{1/2})_3B$, and Li_2Pt_3B is found to be 0.115, 0.16, and 0.105 mΩ cm, respectively, and their respective ρ_{300}/ρ_{10} is 1.3, 1.1, and 1.2. Accordingly, we consider all the above-mentioned arguments concerning the ineffectiveness of the scattering channels in quenching the triplet component to be valid for the case of the solid solution studied in this work.

The specific heat measurements were carried out on two different setups: a heat-pulse relaxation-type calorimeter operating within the range $0.4\text{ K} < T < 20\text{ K}$ and $H \leqslant 70$ kOe, (a commercial one from Oxford Instruments), and a pulse, relaxation-type calorimeter operating within the range $1.8\text{ K} < T < 20\text{ K}$ and $H \leqslant 70$ kOe (a Quantum Design setup).

III. RESULTS AND DISCUSSION

In an earlier work,[12] we argued that $C_e^S(T,0\text{ T})$ of Li_2Pt_3B follows an exponential, activated process and that at very low-temperature, the experimental curve deviates upwards away from this exponential description. Being aware of the difficulty in the stoichiometry control of the highly volatile Li,[2,3] we attributed this deviation to extrinsic effects. Now that there are independent confirmations that such a deviation is intrinsic,[10,11] then, on reanalysis,[11] a quadratic thermal dependence is observed to be a better description (see the inset of Fig. 5 below where the exponential and quadratic descriptions are compared). It is emphasized that due to the above-mentioned variation in the Li stoichiometry, care should be exercised when comparing results from different samples: the reported wide scatter in T_c (H=0) provide the best illustration (see, e.g., Table I). As far as specific heat curves are concerned this is convincingly shown in Fig. 1: though the curves of Fig. 1 are different for $T > T_c$ (H=0), they tend to merge at lower temperatures suggesting that the Li variation is mostly reflected in the lattice contribution. Evidently, only within these lowest temperature regions, the determination of some of the physical parameters (in particular γ) would be sample-independent. At any rate, the overall

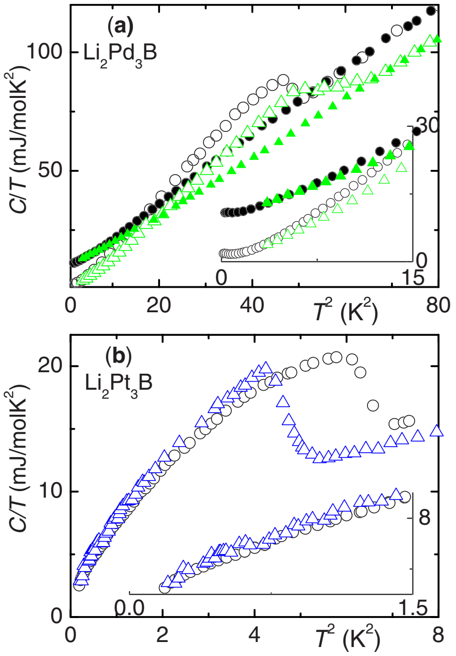

FIG. 1. (Color online) (a) C/T versus T^2 curves of two different samples of Li$_2$Pd$_3$B. Open (closed) symbols represent measurements under zero (▲: 5 T, ●: 7 T) field. (b) Zero-field C/T versus T^2 curves of two different samples of Li$_2$Pt$_3$B measured on two different setups. Data represented by circles (triangles) were measured on a relaxation-(semi-adiabatic-) type calorimeter. In each panel, the inset shows an expanded low-temperature range wherein the two independently measured curves merge (see text).

features of the T-, H-, and x-dependence of these Li-based compounds are found to be the same and as such the conclusions drawn in this work (as well as in other investigations) are almost sample-independent.

A. Normal-state specific heat

The total specific heat of Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B (x=0.0, 0.5, and 1.0) is shown in Figs. 2–4. For the interesting range of temperature (0.4 K$<T<$10 K), the phonon contribution, taken as βT^3, reflects the difference among the atomic weight of Pt and Pd. Since other contributions such as nuclear, crystal-field, or magnetic terms are absent, then the T-, H-, and x-dependence of the specific heat would be shaped exclusively by the electronic degrees of freedom, $C_e(T,H,x)$. Assuming that the phonon contribution is field-independent (at least for the moderate fields employed) then $C_e(T,H,x)$ can be obtained by simple subtraction.

A closer look at the thermal evolution of Figs. 2–4 reveals that for $H>H_{c2}$ the normal-state specific heat evolves as $C_e^N(T,H>H_{c2})=\gamma T+\beta T^3$. Moreover, the insets of Figs. 2–4 show that there are at least two separate regions of temperature, each with a different set of γ and β. For all compounds, the slope of the high-regime linear C/T versus T^2 curve is larger than that of the low-regime one: such a change of slope in the immediate neighborhood of T_c is manifested in many compounds, e.g., the A15 Nb$_3$Sn samples[14] (some aspect of this feature can be detected in the nonmagnetic quaternary borocarbides[15,16]).

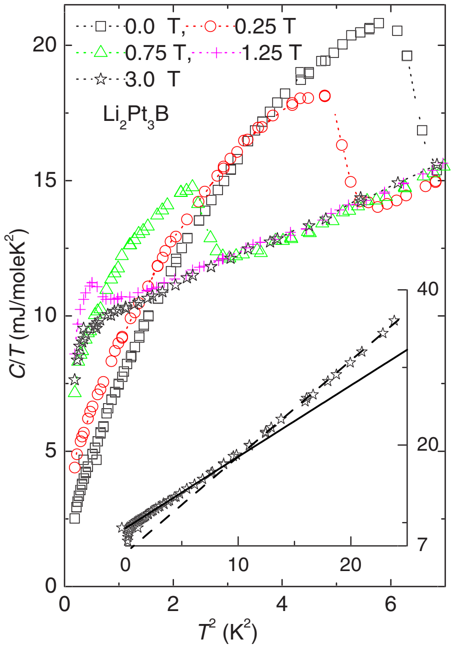

FIG. 2. (Color online) Field-dependent C/T versus T^2 curves of Li$_2$Pt$_3$B. Inset: an extended C/T versus T^2 curve at H=3 T ($>H_{c2}$). The solid (dashed) line represents the fit for $T<T_c$ ($T>T_c$) (see Table II).

It is observed that, on cooling to temperatures below 0.5 K, $C_e^N(T,H>H_{c2})$ of all samples deviates weakly downwards away from this relation; our present experimental conditions do not permit a further clarification. We observed no evidence of field-dependence in γ or β for $T>$0.5 K. Finally, Table II shows that, within the experimental errors, while the values of β do show a consistent x-dependence, those of γ do not show a clear variation, at least not as the trend expected from the electronic structure calculations.[17,18] We note that while the above-mentioned Li nonstoichiometry still poses a problem, however, our present determination of these parameters is more reliable since the measurements had been made on better samples (judged by Figs. 2–4) and extended to a very low temperature range.

B. Superconducting specific heats

The evolution of $C_e^S(T,H)$ of Li$_2$(Pd$_{1-x}$Pt$_x$)$_3$B (x=0.0, 0.5, and 1.0) is shown in Figs. 5–7: as evident, $C_e^S(T,H)$ of the Pd limit shows an exponential dependence (Fig. 7) while that of each of Li$_2$(Pd$_{0.5}$Pt$_{0.5}$)$_3$B (inset of Fig. 6) and Li$_2$Pt$_3$B (inset of Fig. 5) follows a quadratic temperature dependence. Though the normal-state γ (Table II) does not manifest an x-dependence, however, the ratio $\Delta C_e/\gamma T_c$ *does* show that, giving approximately 1.7, 1.1, and 0.8 for x=0.0, 0.5, and 1.0, respectively: this is in good agreement with what is expected when the strength of the ASOC is increased. It is interesting that the quadratic dependence is manifested in $C_e^S(T)$ of Li$_2$(Pd$_{0.5}$Pt$_{0.5}$)$_3$B (see the inset of Fig. 6) revealing that the triplet component of the order parameter is not being

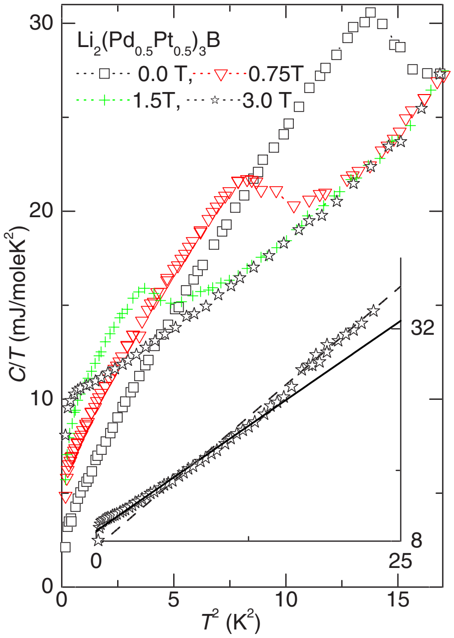

FIG. 3. (Color online) Field-dependent C/T versus T^2 curves of $Li_2(Pd_{0.5}Pt_{0.5})_3B$. Inset: an extended C/T versus T^2 curve at $H=3$ T ($>H_{c2}$). The solid (dashed) line represents the fit for $T<T_c$ ($T>T_c$) (see Table II).

destroyed (see the earlier arguments in the Introduction) and that the line nodes *do* survive the 50% Pd dilution. It is not a mere coincidence that the ratio of the coefficients of the quadratic terms in $Li_2(Pd_{0.5}Pt_{0.5})_3B$ and Li_2Pt_3B, $\alpha_{\mathrm{Pt}_{0.5}\mathrm{Pd}_{0.5}}/\alpha_{\mathrm{Pt}} \simeq 0.63$, is equal to the ratio of the strength of the involved ASOC given by the ratio $Z^2_{\mathrm{Pt}_{0.5}\mathrm{Pd}_{0.5}}/Z^2_{\mathrm{Pt}}=0.63$. As such it does confirm that the quadratic behavior observed in $C_e^S(T,H)$ is driven by the ASOC and lends extra support to the above-mentioned interpretation of the almost linear x-dependence of T_c, H_{c1}, H_{c2}, and $(\partial H_{c2}/\partial T)_{T_c}$ of the series $Li_2(Pd_{0.5}Pt_{0.5})_3B$.

Figure 5 for Li_2Pt_3B and Fig. 6 for $Li_2(Pd_{0.5}Pt_{0.5})_3B$ demonstrate that, in moderate fields, the quadratic T-dependent expression is maintained over a reasonable range of temperatures. This is in accord with the studies of Yuan *et al.*[11] and Nishiyama *et al.*[10] wherein moderate fields do not destroy the line nodes features. Furthermore, when H is increased, $\alpha(H)$ suffers a weak decrease: this may be attributed to a decrease in the condensate contribution. It is noted that for $H>H_{c1}$, the intercept of the $C/\gamma T$ versus T/T_c curve is increased with H: the quasiparticles excitations within the Abrikosov state contribute an additional linear term expressed normally as $\gamma(H)T$; the number of these excitations is proportional either to H or $\sqrt{H}$ depending on the pairing symmetry (see, e.g., Refs. 19 and 20): indeed for the case of the S-wave Li_2Pd_3B superconductor, we observed that at low temperature $\gamma(H)-\gamma(H=0)=\gamma_N H/H_{c2}$. Then, as can be inferred from this observation and verified in Fig. 7, the zero-field exponential character of C_e^S of Li_2Pd_3B is not maintained in any of the applied fields: since the character of such a field-dependent contribution is nonexponential, then the total contribution would not show the Arrhenuis-type behavior, in particular at

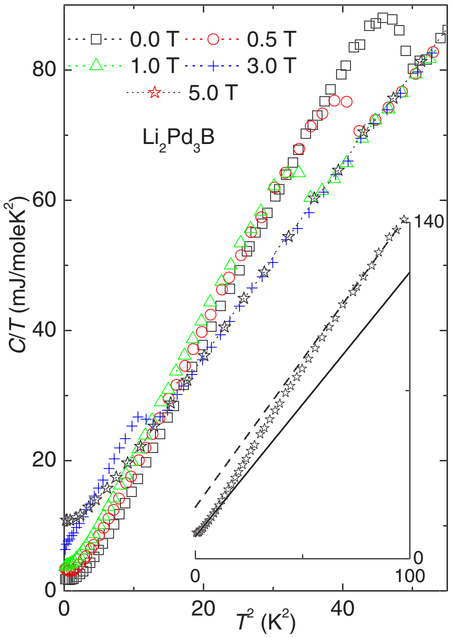

FIG. 4. (Color online) Field-dependent C/T versus T^2 curves of Li_2Pd_3B. Inset: an extended C/T versus T^2 curve at $H=3$ T ($>H_{c2}$). The solid (dashed) line represents the fit for $T<T_c$ ($T>T_c$) (see Table II).

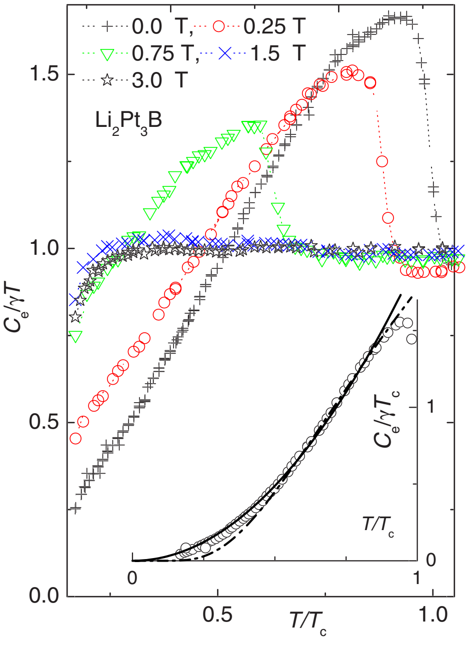

FIG. 5. (Color online) Field-dependent $C_e/\gamma T$ versus T/T_c curve of Li_2Pt_3B. Inset: the zero-field $C_e/\gamma T_c$ versus T/T_c curve. The solid (dashed) line represents the relation $C_e/\gamma T_c=2.0(T/T_c)^2$ [$C_e/\gamma T_c=6.93\exp(-1.37T_c/T)$ mJ/mol K] evidently for $T<T_c$, the calculated quadratic (exponential) curve describes the measured electronic contribution very well in the lower (higher) temperature range. As far as the low-temperature behavior is concerned, the quadratic evolution is a much better description.

TABLE II. The zero-field superconducting specific heat contribution is fitted either to a quadratic term $[C_s(T,0\ \mathrm{T})=\alpha T^2]$ or an exponential term $[C_s(T,0\ T)=\eta\exp(-\Delta/T)]$. γ and β (used in the subtraction of the normal-state contribution) are obtained from the fits shown in Figs. 2–4.

	T_c	γT	βT^3	$C_s(0\ \mathrm{T})=\eta\exp(-\Delta/T)$		$C_s(0\ \mathrm{T})=\alpha T^2$
Li_2T_3B	(K)	γ (mJ/mol K^2)	β (mJ/mol K^4)	η (mJ/mol K)	Δ (K)	α (mJ/mol K)
Li_2Pd_3B	6.95(5)	9.3(3)	1.1(1)	3000	16.4	
$Li_2(Pd_{0.5}Pt_{0.5})_3B$	3.9(1)	9.1(2)	0.95(3)			4.6
Li_2Pt_3B	2.56(5)	9.3(2)	0.92(1)			7.25

very low temperatures where such a field-dependent contribution is relatively larger. It is taken that such type of arguments can be applied to distinguish and clarify the differences between a predominantly spin-singlet, a predominately spin-triplet, or an admixture of spin-triplet and spin-singlet states.

The x-dependent evolution in the specific heats of $Li_2(Pd_{1-x}Pt_x)_3B$ can be better discussed if the zero-field contributions are rewritten as $C_e^S=\alpha(H,x)T^2$ and $C_e^S=\eta(H,x)\exp[-\Delta(H,x)/T]$. It is noted that for both Li_2Pt_3B (Fig. 5) and $Li_2(Pd_{0.5}Pt_{0.5})_3B$ (Fig. 6), $\alpha(H,x)$ increases monotonically with x: an increase in x would increase the strength of the ASOC and as a consequence increase the line-nodes contribution. On the other hand, the strong reduction of ASOC in Li_2Pd_3B (Fig. 7) is conducive to a dominant gapped spin-singlet state with an effective value as given in Fig. 7. Finally, let us discuss the influence of nonmagnetic impurities on the nonconventional superconductivity of Li_2Pt_3B; such impurities are expected to act as depairing centers and consequently lead to a suppression of T_c and to, e.g., a linear-in-T specific heat term at low temperature.[8] That such features are not evident in the superconductivity of $Li_2(Pd_{1-x}Pt_x)_3B$ (see the results of this work and those of Badica *et al.*[3]) confirms our earlier arguments that the alloying process should be considered (not as a depairing effect, but rather) as a means of varying the ASOC.

IV. CONCLUSION

The specific heat of $Li_2(Pd_{1-x}Pt_x)_3B$ ($x=0.0$, 0.5, and 1.0) has been studied as a function of temperature, magnetic field, and Pt concentration. It is found that $C_e^S(T)$ of Li_2Pt_3B and $Li_2(Pd_{0.5}Pt_{0.5})_3B$ follows a quadratic relation revealing an existence of line nodes in the gap function. On the other hand, $C_e^S(T)$ of Li_2Pd_3B follows a conventional exponential behavior suggestive of a nodeless gap function. It is noted that for

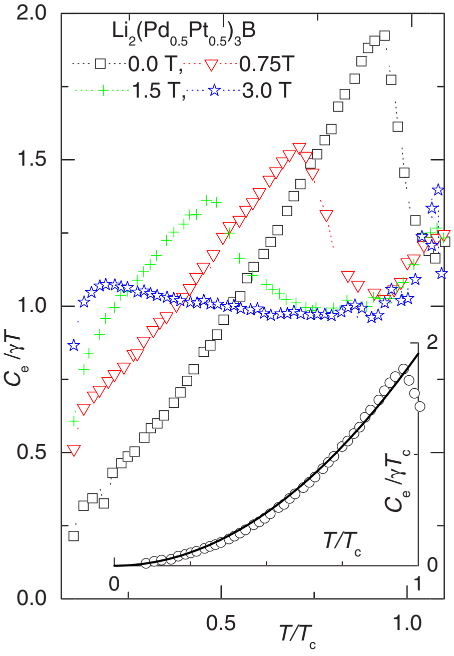

FIG. 6. (Color online) Field-dependent $C_e/\gamma T$ versus T/T_c curve of $Li_2(Pd_{0.5}Pt_{0.5})_3B$. Inset: the zero-field $C_e/\gamma T_c$ versus T/T_c curve. The solid line represents the relation $C_e/\gamma T_c=1.9(T/T_c)^2$. The low-temperature weak deviation from the quadratic behavior may be attributed to minor normal phases.

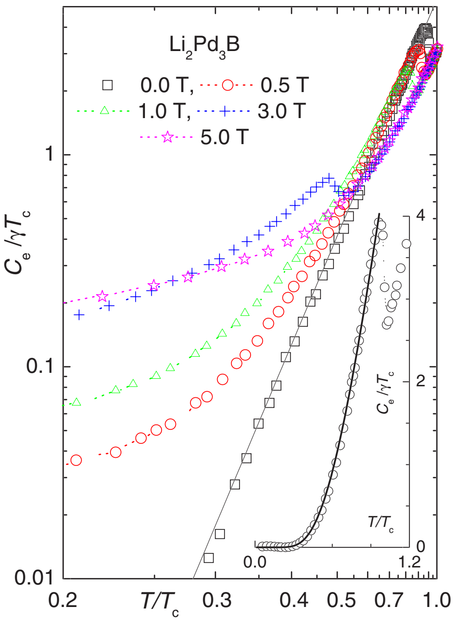

FIG. 7. (Color online) $C_e/\gamma T_c$ versus T/T_c curves of Li_2Pd_3B. To show the exponential character, the abscissa is displayed as a reciprocal of T/T_c while the ordinate is in logarithm base 10. Inset: zero-field $C_e/\gamma T_c$ versus T/T_c curve of Li_2Pd_3B. For $T<T_c$, the solid lines represent $C_e/\gamma T_c=46.4\exp(-2.36T_c/T)$.

both Li_2Pt_3B and $Li_2(Pd_{0.5}Pt_{0.5})_3B$, the coefficient of the quadratic contribution $\alpha(H,x)$ decreases with H but increases with x: an increase in x strengthens the ASOC and as such would increase the contribution from the line nodes. For Li_2Pd_3B, the H- and x-dependent modification of $C_e^S(T,H)$ is as expected for a predominately conventional spin-singlet superconductor.

Evidently, the present specific heat results on these noncentrosymmetric superconductors are in accord with the recent NMR and penetration depth studies and as such constitute an additional confirmation of the statement that the spin-triplet (spin-singlet) state is dominant in the Pt-based (Pd-based) compound. Interestingly, the 50% substitution of the Pt atoms by the lighter Pd atoms does not suppress completely the strength of the ASOC; rather the spin-triplet state is still the dominant one.

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