



Inelastic quasiparticle scattering and T_1^{-1} NQR relaxation rate in $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$

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We present an analysis of recent [M. Yogi *et al.*, J. Phys. Soc. Jpn. **75**, 124702 (2006)] Sb-NQR measurements of the heavy-fermion superconductor $\text{PrOs}_4\text{Sb}_{12}$ and the related La-doped series of superconductors $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$. We find that this data, along with previous temperature-dependent superfluid density results and are well described by a strong-coupling order parameter containing a doping-dependent proportion of the first cubic harmonic ($k_x^4+k_y^4+k_z^4$) component along with an s -wave component, when inelastic scattering due to Coulomb effects are taken into account. The resulting gap function has deep minima along the cubic axis [100], etc. but no nodes. We use the order parameter fits to generate, within the Ginzburg–Landau theory, the effective thermal mass enhancement relative to the $\text{LaOs}_4\text{Sb}_{12}$ material and find that it rapidly varies for small x .

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I. INTRODUCTION

The heavy-fermion superconductor $\text{PrOs}_4\text{Sb}_{12}$, discovered in 2002 by Bauer *et al.*,¹ continues to present substantial difficulties despite a large number of experiments that have since been conducted. Still controversial, for example, are the number and nature (conventional or unconventional) of superconducting phases, the parity (singlet vs triplet) and symmetry of the order parameter, and the source of the pairing interaction in this fascinating material. In particular, a hypothesized second, low-temperature, low-field phase² observed in many samples may well not be intrinsic³ but a final consensus has not been reached.

Given this difficult situation, the best approach seems to be a direct analysis of the experimental data. Recently, Yogi *et al.*⁴ conducted an Sb-NQR relaxation rate (T_1^{-1}) study of the series of superconductors $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$, for $x=0, 0.05, 0.2, 0.4, 0.8$, and 1. $\text{LaOs}_4\text{Sb}_{12}$ is generally accepted to be a weak-coupling phonon-mediated conventional BCS superconductor with an s -wave pairing symmetry, and this fact has important consequences for our analysis. In particular, if $\text{PrOs}_4\text{Sb}_{12}$ were of the triplet type, the addition of the dopant La atoms would be expected to have a drastic effect on T_c , in line with the Abrikosov–Gor’kov theory⁵ of pair-breaking impurities in the unitary limit, as is often invoked for heavy-fermion superconductors.⁶ However, this is not observed, as depicted in Fig. 1; $T_c(x)$ (from Yogi *et al.*⁴) shows a steady, monotonic decrease from the pure Pr compound to the pure La compound. These results found a possible explanation in terms of inelastic scattering on quadrupolar crystalline electric field (CEF) excitations of Pr,⁷ which suggests that $\text{PrOs}_4\text{Sb}_{12}$ is a singlet superconductor. Within the Eliashberg theory used in Ref. 7, an isotropic s -wave gap function was assumed. However, we will show that this model is too simplified to explain the T_1^{-1} results of the $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ series. A considerable anisotropy of the gap has to appear when moving from the La to the Pr side, in conjunction with the mass enhancement, the T_c increase, and the more pronounced strong-coupling effects. However, since the T_c increase is smooth, the order parameter will remain fully symmetric (A_{1g}) for all $x \leq 1$. Given the essentially cubic

symmetry,⁸ this means⁹ that the order parameters $\Delta(\mathbf{k})$ can be expanded in fully symmetric cubic harmonics.¹⁰ The latter can easily be constructed by considering even-parity polynomials in k_x, k_y , and k_z , which are invariant under any permutation of the coordinates. The first cubic harmonic is the s -wave term 1 and the next is $k_x^4+k_y^4+k_z^4$, and we restrict ourselves to these two terms. A similar $s+g$ -wave superconductivity in the tetragonal borocarbides was considered by Yuan *et al.*^{11,12} In general, these terms may be combined to yield a doping dependent $\Delta(\mathbf{k})$ as follows:

$$\Delta(\mathbf{k}) = \alpha(g)\Delta_0[1 - g(k_x^4 + k_y^4 + k_z^4)]. \quad (1)$$

Here, Δ_0 is the Fermi surface maximum of the order parameter, g is the doping-dependent weight of the indicated cubic harmonic (we will refer to this in the rest of the paper as the cubic harmonic), and $\alpha(\equiv \frac{1}{1-g/3})$ is an overall normalization constant chosen to ensure that the maximum value of $\Delta(\mathbf{k})$ is, in fact, Δ_0 .

This choice of order parameter is consistent with the field-angle resolved specific heat data of Custers *et al.*,^{13,14} who found a clear fourfold oscillation pattern in the magnetic fields below H_{c2} , corresponding to the gap minima along all faces of the cube. The oscillation amplitude also substan-

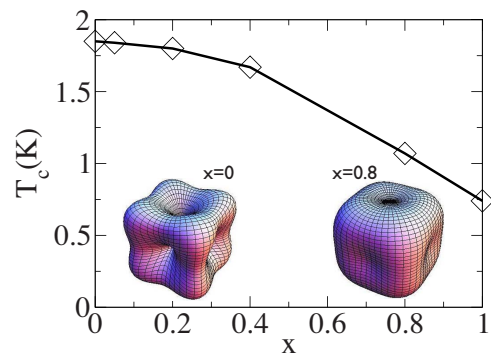


FIG. 1. (Color online) $T_c(x)$ for $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ from Ref. 4. Inset: radial plots of the order parameter for $x=0$ ($g=0.8$) and $x=0.8$ ($g=0.4$).

tially decreased at lower fields $H < 0.3H_{c2}$, indicating that the order parameter does not possess nodes in these directions, in contrast to the results of Izawa *et al.*²

In this paper, we show that all of the T_1^{-1} data generated by Yogi *et al.*⁴ are consistent with the order parameters of the form of Eq. (1), when the effect of inelastic Coulomb scattering is taken into account. This inelastic scattering is of great importance for our analysis. As shown in Ref. 15, in the pure BCS limit, *all* clean-limit anisotropic superconductors are expected to show a small peak in T_1^{-1} just below T_c , generally of a height between 3% and 15% greater than the normal state value at T_c . However, the data of Yogi *et al.*⁴ for $x \leq 0.4$, and indeed most data on anisotropic superconductors, do not show this peak. We demonstrate below that this can be understood as a consequence of inelastic quasiparticle scattering, which can be expected to be most prominent right below T_c , and then rapidly decreases thereafter, becoming exponentially activated as $T \rightarrow 0$.

We use the existence or absence of this peak in various data as a strong indicator of the magnitude of the cubic harmonic, given that pure *s*-wave superconductors generally show substantial peaks, while strongly anisotropic superconducting materials generally do not. Our strategy is to use g as the primary fitting parameter in what follows; in general, this work uses a minimum of fitting parameters.

II. CALCULATION

The standard expression for T_1^{-1} is as follows:

$$(T_1 T)^{-1} = (T_1 T)_{T_c}^{-1} \int_0^\infty \frac{dE}{2T} [N^2(E) + M^2(E)] \operatorname{sech}^2\left(\frac{E}{2T}\right). \quad (2)$$

Here, E is the quasiparticle energy, and $N(E)$ and $M(E)$ are the quasiparticle density of states (DOS) and ‘‘anomalous’’ DOS, respectively, given in the BCS approximation by

$$N(E) = \left\langle \frac{E}{\sqrt{E^2 - \Delta(\mathbf{k})^2}} \right\rangle_{FS}, \quad (3)$$

$$M(E) = \left\langle \frac{\Delta(\mathbf{k})}{\sqrt{E^2 - \Delta(\mathbf{k})^2}} \right\rangle_{FS}. \quad (4)$$

Here, we assumed a spherical Fermi surface so that the relevant expectation value means $\int \frac{d\Omega}{4\pi}$. This assumption is justified since the largest Fermi surface¹⁶ of $\text{PrOs}_4\text{Sb}_{12}$ consists of electron spheroids centered around the H -points, e.g., $(\pm 2\pi, 0, 0)$, within a nearest neighbor tight-binding approximation. (Note the use of the hole picture in Ref. 16.) From dHvA experiments, it is also known that the Fermi surface of the Pr compound ($x=0$) is virtually identical to that of the La compound ($x=1$) (Ref. 17) so that one may expect that the same holds true for intermediate x .

Ordinarily, in such T_1^{-1} computations, one neglects the quasiparticle lifetime effects. However, as stated earlier, this results in a peak in T_1^{-1} just below T_c , for all x and all g . This peak is not observed for $x \leq 0.4$. We, therefore, account for such effects by allowing the quasiparticle energy E in the

DOS calculations to have a small imaginary, temperature-dependent part $\Gamma(T)$, as was introduced by Dynes *et al.*,¹⁸ such that $\tilde{E} = E - i\Gamma(T)$.

Rather than simply choosing this $\Gamma(T)$ as a temperature-dependent fitting parameter for each of the six doping levels studied by Yogi *et al.*, we adopt a unified approach for *calculating* $\Gamma(T)$. In this, the complete set of $\Gamma(x, T)$ has only *one* fitting parameter, which sets the overall scale of the quasiparticle decay rate for all doping levels. The only other fit parameters in the entire T_1^{-1} calculation are the weight g of the cubic harmonic, which, of course, affects the regular and anomalous DOS, and the Δ_0/T_c ratios for $x \leq 0.4$.

This unified approach is based upon the work of Devereaux and Belitz,¹⁹ which described the effect of disorder upon inelastic Coulomb quasiparticle scattering. This work was performed for conventional, *s*-wave superconductors, and here, we modify this for the current situation, as described below. Although it is well known²⁰ that, in conventional clean superconductors, it is the electron-phonon interaction that gives the strongest contribution to inelastic scattering, Devereaux showed that in disordered superconductors (a reasonable model considering the variable doping of La), it is the Coulomb term that is most significant. This arises from diffusion enhancement of the quasiparticle scattering rate,¹⁹ which results in a stronger effective interaction (quantitatively, for three dimensions, the inelastic Coulomb scattering rate is increased by a factor of $\sqrt{E_F/T}$). Devereaux found the inelastic quasiparticle scattering rate to be

$$\Gamma(T) = \Delta \frac{F (3\pi)^{3/2}}{Z 2^{3/2}} (\Delta/E_F)^{1/2} \hat{\rho}^{3/2} (T/\Delta)^{1/2} e^{-\Delta/T}, \quad (5)$$

$$\propto \Delta T^{1/2} \hat{\rho}^{3/2} e^{-\Delta/T}. \quad (6)$$

Here, F is an electrostatic screening factor, which we assume as constant, Z is the real part of the normal self-energy, which we take as constant, and $\hat{\rho} = \rho/\rho_M$, with ρ as the normal state resistivity and $\rho_M = \frac{2\hbar\pi^2}{e^2 k_F}$. We have used Devereaux’s notation here to avoid confusion.

The important point here is the proportionality in the last line of the above equation. Nearly all of the parameters contained here are experimentally accessible and, thus, do not represent free fitting parameters. In particular, the normal state resistivity of $\text{LaOs}_4\text{Sb}_{12}$ (for T just above T_c) is approximately 1/3 that of $\text{PrOs}_4\text{Sb}_{12}$,¹⁷ while its T_c of 0.73 K is approximately 2/5 of the $\text{PrOs}_4\text{Sb}_{12}$ value. Taken together, these mean that in units of Δ_0 (which varies along the series), the quasiparticle scattering rate near T_c , where the exponentially activated factor in $\Gamma(T)$ is of less importance and is approximately eight times smaller in the $\text{LaOs}_4\text{Sb}_{12}$ material. In practice, this means that, within this theory, one can consistently explain both the observed lack of the expected coherence peak for $\text{PrOs}_4\text{Sb}_{12}$ and the substantial (twice the height value at T_c) peak observed in the T_1^{-1} of $\text{LaOs}_4\text{Sb}_{12}$. For the doping levels in between these points, we have assumed a resistivity $\rho(x)$ using the form $\rho(x) = [\rho(0) + \rho(1)]/2 + [\rho(1) - \rho(0)] \cos(\pi x)/2$, which smoothly interpolates between $x=0$ and $x=1$.

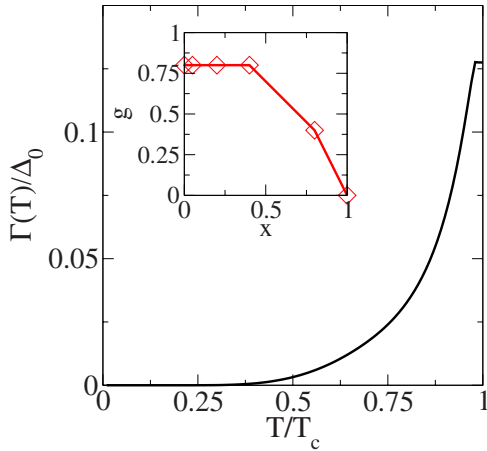


FIG. 2. (Color online) The inelastic scattering rate $\Gamma(T)$ for $\text{PrOs}_4\text{Sb}_{12}$. Inset: the cubic harmonic content g as a function of x .

One point to clarify is that, since the order parameter $\Delta(\mathbf{k})$ for the various doping levels is anisotropic, some further assumption is necessary in order to apply Devereaux's result to our situation. An important issue, detailed in the next section, is that none of the T_1 results can be fit using an order parameter that contains point nodes, as was assumed in Ref. 9. The decline in T_1^{-1} below T_c for $\text{PrOs}_4\text{Sb}_{12}$ is far too rapid for an order parameter that contains nodes and, in addition, the superfluid density²¹ for low T is not fit by the model of Parker *et al.*⁹ This means that the quasiparticle inelastic decay rate is effectively “gapped,” so that we simply replace Δ in the Devereaux formula by its Fermi surface average. We note that the magnitude of this averaged order parameter depends on temperature and x . A plot of the inelastic scattering rate Γ , thus employed for $\text{PrOs}_4\text{Sb}_{12}$, is shown.

As noted, we have also included a superfluid density calculation for the pure Pr ($x=0$) case, which we evaluated via the standard expression (assuming an order parameter that retains cubic symmetry, so that the ρ tensor is isotropic) (Ref. 22),

$$\rho_s(T) = 1 - \int_0^\infty \frac{dE}{2T} N(E) \text{sech}^2\left(\frac{E}{2T}\right). \quad (7)$$

III. RESULTS

Depicted in Fig. 3 is the main result of this paper. We have plotted the observed T_1^{-1} with our best fits using the methodology described above and found excellent agreement for all six doping values x . We note that the T_1^{-1} results for $x=0$, $x=0.05$, and $x=0.2$ essentially fall on top of one another, and for these three dopings, we have taken $g=0.8$. A significantly larger value would imply second order nodes, in contradiction to the data of Custers *et al.*,¹³ while a significantly smaller value would generally produce a peak in T_1^{-1} , which were not observed for these dopings. A plot of $g(x)$ is contained in the inset of Fig. 2. The $g=0.8$ fit means that these materials contain a minimum Fermi surface excitation energy of approximately $0.25\Delta_0$, thus producing exponentially activated behavior at low energy. This is consistent

TABLE I. The doping levels, fit parameters, and weak-coupling Δ_0/T_c values.

Doping (x)	Parameters		
	g	Δ_0/T_c	$(\Delta_0/T_c)_{wc}$
0.0	0.8	3.78	2.26
0.05	0.8	3.78	2.26
0.2	0.8	3.78	2.26
0.4	0.8	2.78	2.26
0.8	0.4	1.99	1.99
1.0	0.0	1.76	1.76

with the field-angle resolved specific heat measurements^{13,14} for $x=0$. They suggest deep minima of the gap amplitude along the three cubic axes. The relative angular oscillation amplitude, however, drops to zero for a small field, indicating a small but fully formed gap along these directions.

We also note that the small feature at T_c is essentially absent for these order parameters, matching the experiment. In order to attain a T_1^{-1} , which falls sufficiently rapidly below T_c , it was necessary to employ a strong-coupling ratio $\Delta_0/T_c=3.78$, which significantly exceeds the weak-coupling value of 2.26 for this order parameter. Experimental Δ_0/T_c values from three $\text{PrOs}_4\text{Sb}_{12}$ specific heat studies^{23–25} range from 2.6 to 3.7, so our value is reasonable. In addition, $\Delta(T)$ was taken to have a strong-coupling type temperature dependence, with relatively little reduction in its zero-temperature value approximately below $0.8T_c$. We note that the $x=0.4$ data significantly deviates from the data of the previous three dopings; it does not drop nearly as rapidly below T_c (Table I), although it, too, lacks a peak just below T_c . This lack of a peak mandates a large cubic harmonic component and, accordingly, we have chosen $g=0.8$ here as well. However, we have treated this material near the weak-coupling limit, so that T_1^{-1} does not drop as rapidly near T_c . This is appropriate

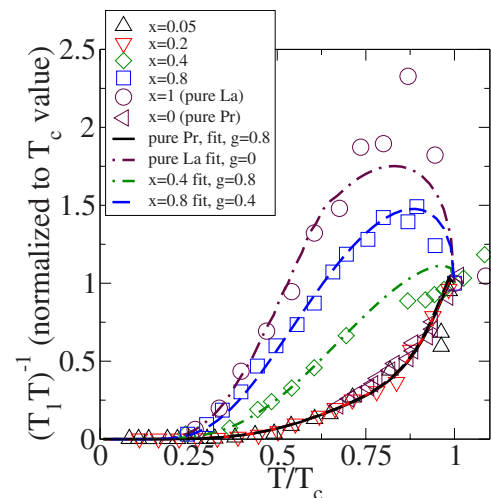


FIG. 3. (Color online) The experimental data and theoretical T_1^{-1} curves for $x=0$, 0.05, 0.2, 0.4, 0.8, and 1.0, as indicated. Residual lifetime broadening $\Gamma=0.01 \Delta_0$ (Ref. 18) used for $x=1$ curve for $T < 0.6T_c$.

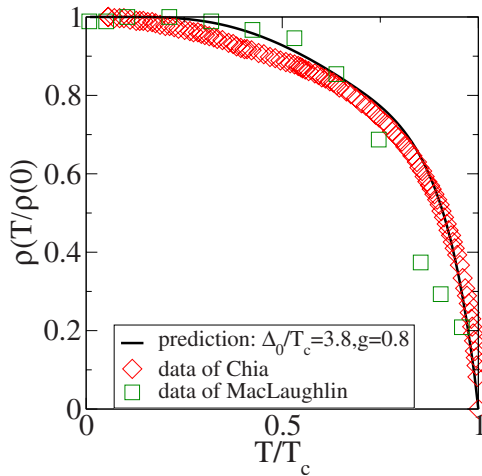


FIG. 4. (Color online) The fit to the superfluid density data of $\text{PrOs}_4\text{Sb}_{12}$. Same parameters as in Fig. 3.

given our later discussion of the effective masses of the $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ series, where we show a substantial effective mass enhancement only for $x < 0.4$, suggesting that correlation effects are minimal at this point and indicating a weak-coupling analysis. We note that the theory predicts a small peak in T_1^{-1} just below T_c , which is not observed; we attribute this to the relative simplicity of our treatment of the inelastic Coulomb scattering.

Both the $x=0.8$ and $x=1$ (pure La) data show substantial T_1^{-1} peaks below T_c , and the calculation reproduces these relatively well, with the exception of one outlying data point for the pure La case. The low-temperature behavior is also reproduced relatively well. The cubic harmonic component g , a fitting parameter, varies relatively rapidly between these two dopings, taking the value 0.4 for $x=0.8$ but 0 for $x=1.0$. We believe that this indicates the effective strength of the Pr substitution in pushing the system toward a nodal superconducting state.

By using the same parameters as in Fig. 3, Fig. 4 depicts a fit of the superfluid density data of Chia *et al.*²¹ for $\text{PrOs}_4\text{Sb}_{12}$. We note excellent agreement, particularly in the region near T_c , whose virtually vertical slope has, so far, defied theoretical interpretation. Mean-field theories generally produce $\rho(T \rightarrow T_c)$, which linearly vanish at T_c , usually with a slope between one and two. Our ρ has a slope of approximately six at T_c as a result of the very strong-coupling nature of this material. For completeness, we also show the superfluid density obtained from the muon spin rotation data of MacLaughlin *et al.*,²⁶ which shows somewhat different behavior near T_c and at low temperature. Our prediction deviates from Chia's data mostly at low temperature, where it is exponentially activated, due to the small gap where $\Delta(\mathbf{k})$ has its minimum at approximately $0.25\Delta_0$. Chia's low-temperature data are consistent with power-law behavior, while that of MacLaughlin more closely approaches the activated behavior.

IV. GINZBURG-LANDAU THEORY AND EFFECTIVE MASS ENHANCEMENT

As a final application of the order parameters determined by the fits to the T_1^{-1} data, we determine the increase in

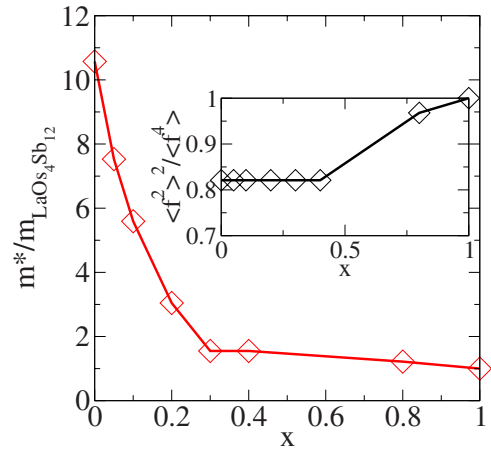


FIG. 5. (Color online) The effective thermal mass in the $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ series, adapted from the data of Ref. 28. Inset: the $\frac{\langle f^2 \rangle^2}{\langle f^4 \rangle}$ ratio.

effective mass that occurs as the pure Pr compound is approached. The jump in specific heat ΔC at T_c can be expressed in terms of the 2nd and 4th order Ginzburg-Landau²⁷ coefficients α and β as

$$\Delta C = T_c \frac{\alpha^2}{\beta}, \quad (8)$$

while the normal state specific heat at T_c can be written as

$$C_n(T_c) = \frac{2\pi^2}{3} T_c N(E_F). \quad (9)$$

Given an order parameter $f(\mathbf{k})$ normalized to unity as its maximum value [i.e., $\Delta(\mathbf{k}) = \Delta(T)f(\mathbf{k})$], one has the following relationship from the BCS weak-coupling theory:²²

$$\frac{\Delta C}{C_n} = \frac{12}{7\zeta(3)} \frac{\langle f^2 \rangle^2}{\langle f^4 \rangle}, \quad (10)$$

where $\zeta(3) (=1.202, \dots)$ is the Riemann zeta function and the relevant averages are over the Fermi surface. Although strictly speaking, we have deviated from the weak-coupling approach in our Δ/T_c ratios, as a qualitative estimate this expression remains valid. Using the expression

$$N(E_F) = \frac{m^* k_F}{\hbar^2 \pi^2}, \quad (11)$$

one finds

$$\frac{\Delta C}{T_c} = \frac{8k_F}{7\zeta(3)\hbar^2} m^* \frac{\langle f^2 \rangle^2}{\langle f^4 \rangle} \equiv a_0 m^* \frac{\langle f^2 \rangle^2}{\langle f^4 \rangle}. \quad (12)$$

Now, the relevant point is that the prefactor a_0 in the above equation depends only on k_F , which is the same in the entire range of doped compounds $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$, as the Pr and the La donate the same number of electrons to the system, and dHvA results¹⁷ indicate nearly equal cross-sectional Fermi surface areas for the $x=0$ and $x=1$ materials. This means that once one corrects the effects of the anisotropic order parameters, one may extract the effective thermal mass from specific heat data.^{28,29} We have plotted this mass in Fig.

5, relative to the mass in $\text{LaOs}_4\text{Sb}_{12}$. It is immediately apparent that there is a substantial mass enhancement for $x < 0.3$, which corresponds to the region where the superconductivity is most enhanced from the weak-coupling regime. For $x=0.4$, the fitted $\Delta(0)/T_c$ ratio is 2.78, which is nearer to the weak-coupling value 2.26 for this order parameter (here, $g=0.8$) than the lower dopings. Thus, it appears that the effective mass enhancement and strong-coupling superconductivity may have a common origin. Indeed, this has also been the conclusion from the microscopic model of Ref. 7, based on quadrupolar CEF excitations of Pr.

V. CONCLUSION

To conclude, we have shown that the T_1^{-1} data on the $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ series can be consistently explained by an

order parameter model combining an s -wave and a doping-variable cubic harmonic component by taking into account the influence of inelastic Coulomb scattering upon the normal and anomalous DOS. We further find that this model fits the superfluid density data of $\text{PrOs}_4\text{Sb}_{12}$ well and that the dopings $0 \leq x \leq 0.2$, which are best fit by strong-coupling calculations also show substantial effective mass enhancement.

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