Field dependent mass enhancement in $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ from aspherical Coulomb scattering

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The scattering of conduction electrons by crystalline electric field (CEF) excitations may enhance their effective quasiparticle mass similar to scattering from phonons. A wellknown example is Pr metal where the isotropic exchange scattering from inelastic singlet-singlet excitations causes the mass enhancement. An analogous mechanism may be at work in the skutterudite compounds $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ where close to $x=1$ the compound develops heavy quasiparticles with a large specific heat $\gamma$. There the low lying CEF states are singlet ground state and a triplet at $\Delta = 8$ K. Due to the tetrahedral CEF the main scattering mechanism must be the aspherical Coulomb scattering. We derive the expression for mass enhancement in this model including also the case of dispersive excitations. We show that for small to moderate dispersion there is a strongly field dependent mass enhancement due to the field induced triplet splitting. It is suggested that this effect may be seen in $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ with suitably large $x$ when the dispersion is small.

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I. INTRODUCTION

The filled-skutterudite compound $\text{PrOs}_4\text{Sb}_{12}$ has recently obtained considerable attention. There are several reasons for that. It is a heavy fermion ($\gamma \sim 350\sim 500\text{mJ/molK}^2$) superconductor with a transition temperature of $T_c(\text{Pr}) = 1.85$ K. This temperature is larger than the one of the related system $\text{LaOs}_4\text{Sb}_{12}$ which is $T_c(\text{La}) = 0.74$ K. A number of experiments, like those on Sb-NMR relaxation rate in $\text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_{12}$ [1] suggest that the superconducting order parameter is of the conventional isotropic $s$-wave type with possible admixture of higher harmonics depending on the Pr content [2]. However, questions and ambiguities remain. They concern the experimental findings for the penetration depth [3, 4] and initial studies of the thermal conductivity in a rotating magnetic field [5]. For example, the former suggest a possible nodal structure while the latter in addition points towards two distinct superconducting phases. The observed enhancement of the superconducting transition temperature by more than a factor of two when La is replaced by Pr seems surprising at first sight. It is well known that when Pr ions are added as impurities to an $s$-wave superconductor like $\text{LaPb}_3$ it suppresses the superconducting transition temperature rather efficiently. So why does it enhance $T_c$ in the present case? Since the phonons in $\text{LaOs}_4\text{Sb}_{12}$ and $\text{PrOs}_4\text{Sb}_{12}$ are very nearly the same, the enhancement must come from the two $4f$ electrons which $\text{Pr}^{3+}$ has. The heavy fermion behavior of $\text{PrOs}_4\text{Sb}_{12}$ seems puzzling too. It shows up in a large specific heat jump $\Delta C/T_c \approx 500 \text{mJ/(molK}^2$) at $T_c$ and also in a large effective mass in de Haas-van Alphen experiments. The Kondo effect cannot be the origin of the heavy quasiparticles since the $4f^2$ electrons are well localized with a Hund’s rule total angular momentum $J = 4$ and a non-Kramers ground state. The key to the enhancement of $T_c$ and the formation of heavy quasiparticle excitations lies in the crystalline electric field (CEF) splitting of the $J = 4$ multiplet (Sect. III), together with the aspherical Coulomb scattering mechanism of conduction electrons from CEF excitations [6, 7]. The self energy and effective mass enhancement due to this mechanism will be calculated in Sect. III. The CEF states, their excitation energies and matrix elements are modified by an external field. The ensuing effective mass dependence on the field which is the main topic of the present work is calculated in Sect. VI for dispersionless excitations and in Sect. VII for the case with dispersive quadrupolar excitons. Some numerical results are discussed in Sect. VII and Sect. VIII finally gives the conclusions.

II. THE CEF STATES OF Pr IN $T_h$ SYMMETRY AND THEIR INTERACTIONS

From inelastic neutron scattering the CEF energy levels are known. The compound has tetrahedral $T_h$ site symmetry for Pr. The data are explained best by a CEF ground state

$$|\Gamma_1\rangle = \frac{\sqrt{30}}{12} (|+4\rangle + |-4\rangle) + \frac{\sqrt{21}}{6} |0\rangle$$

(1)
FIG. 1: Left (a): Fermi surface of n.n.n. tight binding model in hole representation in the bcc Brillouin zone. Right (b): schematic Fermi surface in electron representation in the 2D projected Brillouin zone. It consists of spheroids around the equivalent H-points \((\frac{2\pi}{a}, 0, 0)\). The polar angle \(\theta\) of \(q\) is given by \(\theta = \frac{1}{2}(\pi - \theta')\). Furthermore we have \(q = 2p_F \sin \theta' = 2p_F \cos \theta\) where \(p_F\) is the Fermi momentum. The geometric restrictions require \(0 \leq q \leq 2p_F\) and \(0 \leq \theta \leq \frac{\pi}{2}\).

with a low-lying triplet excited state at an energy of \(\Delta = 8K\) [6, 8, 9, 10]. The other CEF levels are so high in energy that they can be neglected. The \(\Gamma_t\) triplet state of \(Th\) symmetry is a superposition of two triplets \(\Gamma_4\) and \(\Gamma_5\) of \(Oh\) symmetry. More specifically one finds [9, 10]

\[
| \Gamma_t, m \rangle = \sqrt{1 - d^2} | \Gamma_5, m \rangle + d | \Gamma_4, m \rangle , \quad m = 1...3
\]  

with states of \(Oh\) symmetry given by

\[
| \Gamma_5, \pm \rangle = \pm \sqrt{\frac{7}{8}} | \pm \rangle \mp \sqrt{\frac{1}{8}} | \mp \rangle \quad | \Gamma_5, 0 \rangle = \sqrt{\frac{1}{2}} (| +2 \rangle - | -2 \rangle) \\
| \Gamma_4, \pm \rangle = \mp \sqrt{\frac{1}{8}} | \mp \rangle \mp \sqrt{\frac{7}{8}} | \pm \rangle \quad | \Gamma_4, 0 \rangle = \sqrt{\frac{1}{2}} (| +4 \rangle - | -4 \rangle) 
\]  

(3)

The conduction electrons interact with the CEF energy levels of the Pr\(^{3+}\) ions. The most important ones are the isotropic exchange interactions and the aspherical Coulomb scattering. The former is of the form

\[
H_{ex}(i) = -2 (gJ - 1) J_{ex} \sum_{kq\sigma\sigma'} (s_{\sigma\sigma'} \cdot \mathbf{J}_i) c_{k-q\sigma'}^t c_{k\sigma}
\]  

(4)

where \(c_{k\sigma}^t (c_{k\sigma})\) are the creation (annihilation) operators for conduction electron with momentum \(k\) and spin \(\sigma\) while \(s\) is their spin operator. Furthermore \(gJ\) is the Landé factor. The aspherical Coulomb interaction in local orbital basis is given by [11]

\[
H_{AC}(i) = \left( \frac{5}{4\pi} \right)^\frac{1}{2} \sum_{kk'\sigma} \sum_{m=-2}^{+2} I_2(k's; kd) Q_2 \left[ Y_2^m(\mathbf{J}_i) c_{k'\sigma}^t c_{kd\sigma} + h.c. \right]
\]  

(5)
FIG. 2: Static (normalised) quadrupolar susceptibility $\Delta \chi_Q$ as function of magnetic field with $(d^2 = 0.067$ or $|d| = 0.26)$ and without $(d^2 = 0)$ tetrahedral CEF. Full lines correspond to evaluation with Eq. (24) while the dashed line corresponds to the low field expansion Eq. (26). Approaching the $\Gamma_4 - \Gamma_2^0$ level crossing leads to an increasing $\chi_Q$ and mass enhancement which becomes singular at the crossing $h_c/\Delta = (1 - |d|^2)^{-1}$. The latter is pushed to higher field for increasing $d^2$ which reduces the increase of $\chi_Q$. For $d^2 \geq 0.42$ $\chi_Q$ decreases with field strength because the tetrahedral CEF leads to $\Gamma_4 - \Gamma_2^0$ repulsion. The mass enhancement is proportional to the quadrupolar susceptibility with $\delta m^*/m = g_{eff}(\Delta \chi_Q)$ and $g_{eff} = (\tilde{g}/\Delta)f$. For $d^2 = 0.067$ and $g_{eff} = 0.077$ (Sect. V) one has $(\delta m^*/m)_{h=0} = 16$.

Here $c_{k\sigma\nu}$ destroys a conduction electron with momentum $k = |k|$, in a $l = 2$ state with azimuthal quantum number $m$ and spin $\sigma$ and $c_{k\sigma\nu}^\dagger$ creates one with momentum $k'$ in a $l = 0$ state. The Coulomb integral $I_2$ is defined, e.g., in Ref. [11] and $Q_2$ is the quadrupole moment of the Pr$^{3+}$ ion. The operators $Y_{2}^{m}(J)$ are given by

$$
Y_{2}^0 = (2/3)^{1/2} \left[ 3J_z^2 - J(J + 1) \right] / N_J \\
Y_{2}^\pm = \pm \left( J_z J^\pm + J^\pm J_z \right) / N_J \\
Y_{2}^{\pm 2} = \left( J^\pm \right)^2 / N_J
$$

with $N_J = (2/3)^{1/2}(2J^2 - J)$. $H_{AC}$ leads to a transfer of angular momentum $l = 2$ between the conduction electrons and the incomplete $4f$ shell. It is a quadrupolar type of interaction.

An important feature of PrOs$_4$Sb$_{12}$ is the experimental finding that the low-energy triplet state has a small value of $|d| = 0.26$ with the implication that the inelastic scattering of the conduction electrons is predominantly of quadrupolar character. With this information the two features pointed out above, i.e., the increase of $T_c$ when La is replaced by Pr and the heavy quasiparticle mass can be understood quantitatively [8]. As has been known for a long time quadrupolar inelastic scattering of conduction electrons by low-energy CEF levels enhances Cooper pairing since these excitations act similarly as a localized phonon mode. The difference is that phonons are related to changes in the ion position while intra-atomic quadrupolar CEF excitations are related to changes of the $4f^2$ wavefunction.

Also the heavy quasiparticle mass is related to the inelastic scattering processes of the conduction electrons. This feature has been previously exploited to explain the mass enhancement found in Pr metal using the isotropic dipolar exchange interaction $H_{ex}$ [13]. As mentioned above in Pr$_{1-x}$La$_x$Os$_4$Sb$_{12}$ the spherical Coulomb scattering $H_{ac}$ is dominant over exchange scattering. For this model a quantitative calculation of the changes in $T_c$ and the mass enhancement as function of Pr concentration are found in Ref. [7].

The aim of the present communication is to extend the previous calculations by including the effect of an external magnetic field on the mass enhancement. A field splits the triplet states and leads to a decrease of the excitation energy of one of the three states, at least for small tetrahedral admixture $d$ as in PrOs$_4$Sb$_{12}$. Therefore an increase of the effective mass with increasing field is expected in this case.

The situation is different for Pr metal mentioned before where ground state and first excited state are two singlets. There a magnetic field pushes the two energy levels apart and hence increases the excitation energy. As a consequence the effective quasiparticle mass decreases with increasing external field in agreement with experimental findings [14]. In the present singlet-triplet model this case would be realised for $|d| > 0.65$. 
III. SELF ENERGY AND MASS RENORMALISATION

We start out by specifying the electronic part of the Hamiltonian for the system \( \text{La}_{1-x}\text{Pr}_x\text{Os}_4\text{Sb}_{12} \). It is of the type

\[
H = H_{el} + H_{CEF} + H_{AC} + H_{ex} + H_Z .
\]

Here \( H_{el} \) is of the conventional form and need not be explicitely written down. It contains the conduction band dispersion which may be described by a n.n.n. tight binding model \(^{15}\) according to

\[
\epsilon_{k\sigma} = t \cos \frac{1}{2} k_x \cos \frac{1}{2} k_y \cos \frac{1}{2} k_z + t'(\cos k_x + \cos k_y + \cos k_z)
\]

with \( t=174 \text{ meV} \) and \( t'=-27.84 \text{ meV} \). The transfer integrals \( t \) and \( t' \) are chosen so as to reproduce the observed linear specific heat coefficient \( \gamma = 36 \text{mJ/(mole K}^2 \text{)} \) of the non-f reference compound \( \text{LaOs}_{12}\text{Sb}_{12} \) \(^{16}\). In the electron picture the associated Fermi surface consists of H-centered spheroids with a Fermi radius \( p_F \simeq 0.7(2\pi/a) \), see Fig. \( \text{Ib} \). Aside from subtle effects this is quite similar to the LDA FS in Ref. \( 17 \) (note that in this reference the FS is depicted as in Fig. \( \text{Ib} \)). It corresponds to a single band originating in Sb-4p states.

The CEF and Zeeman Hamiltonian are

\[
H_{CEF} + H_Z = \sum_{i,i'=n} E_{\Gamma} |\Gamma_n(i)\rangle \langle \Gamma_n(i)| + g_J \mu_B \sum_i |\mathbf{J}(i)\rangle \langle \mathbf{J}(i)| \mathbf{H} .
\]

The external magnetic field is denoted by \( \mathbf{H} \), \( g_J \) is the Landé factor and \( \mu_B \) is Bohr’s magneton. Furthermore \( i \) labels the Pr sites, and \( |\Gamma_n\rangle \) denotes the singlet ground-state \( |\Gamma_s\rangle \) and the triplet \( |\Gamma_t\rangle \) (see Eq. \( 2 \)) with energies \( E_s = 0 \) and \( E_t = 8 \text{ K} \), respectively. We assume that not only the phonons but also their local interactions with the electrons are independent of partial replacements of La by Pr.

As pointed out before the system has \( T_h \) symmetry but since the CEF transition can be reduced to those between states of cubic symmetry (see \( 2,3 \)) we specialize \(^{15}\) to cubic symmetry. In that case the aspherical Coulomb interaction written in a basis of Bloch states becomes

\[
H_{AC}(i) = g \sum_{kq} \sum_{\alpha\beta\sigma} O_{\alpha\beta}^{\sigma} \hat{q}_\alpha \hat{q}_\beta c_{k\sigma}^\dagger c_{k\sigma} e^{i\mathbf{k}\mathbf{R}_i}.
\]

with \( O_{\alpha\beta} = \frac{\gamma}{2}(J_\alpha J_\beta + J_\beta J_\alpha) \), \( \hat{q}_\alpha = q_\alpha/|\mathbf{q}| \) and \( \alpha\beta = yz, zx, xy \) denoting the three quadrupole operators with \( \Gamma_3 \) symmetry. The remaining \( \Gamma_3 \) quadrupole terms are neglected since they do not couple to the singlet-triplet excitations. The coupling constant \( g \) may in principle be determined by experiments. A derivation of \( 11 \) may be obtained from Ref. \( 18 \).

In order to determine the effective mass \( m^* \) of the quasiparticles at zero temperature one must calculate the Green’s function of conduction electrons.

\[
G(p, \omega) = \frac{1}{\omega - \epsilon(p) - \Sigma(p, \omega)} .
\]

The effective mass enhancement due to interactions of the conduction electrons follows from

\[
\frac{m^*}{m} = 1 - \frac{\partial \Sigma(p_F, \omega)}{\partial \omega} \bigg|_{\omega=0}
\]

where \( p_F \) is the Fermi momentum and \( m \) is the reference quasiparticle mass including band effects and effects of electron-phonon coupling. Neglecting vertex corrections the irreducible electron self energy \( \Sigma(p, \omega) \) due to \( H_{AC} \) is given by

\[
\Sigma(p, \omega) = g^2 \sum_{\alpha\beta,n} \int \frac{d^3 q}{(2\pi)^3} |\Lambda^n_{\alpha\beta}(\mathbf{q})|^2 \int \frac{d\omega'}{2\pi} D_n(\mathbf{q}, \omega') G(p - \mathbf{q}, \omega - \omega') ,
\]
Here \( D_n(\mathbf{k}, \omega) \) denotes the boson propagator of CEF excitations. It is related to the dynamical quadrupolar susceptibility of the CEF system. In the present case we will neglect effective RKKY type interactions between CEF states on different sites therefore the boson propagator is local (\( \mathbf{q} \)-independent). The momentum dependence of the bare vertex \( \Lambda_{\alpha\beta}^n(\mathbf{q}) \) is due to the quadrupolar \( \Gamma_5 \) form factors in Eq. (10). It is defined as

\[
\Lambda_{\alpha\beta}^n(\mathbf{q}) = g\bar{q}_\alpha \bar{q}_\beta (\Gamma_5 |O_{\alpha\beta}| \Gamma_\pi^n) .
\]  

The self energy due to exchange scattering which involves the magnetic susceptibility can be safely neglected because of the smallness of \( d^2 \) (see (2)) as a more detailed investigation including matrix elements and coupling constants shows. The propagator of the local singlet-triplet boson excitations is given by

\[
D_n(\mathbf{q}, \omega) = D_n(\omega) = \frac{2\delta_n}{\delta_n^2 - \omega^2} .
\]  

Here the field dependent singlet-triplet excitation energies are given by \( \delta_{n}(H) = \epsilon_{r}(H) - \epsilon_{s}(H) \) (n = +,0,−).

The self-energy in Eq. (13) can be evaluated following Migdal’s integration procedure (see, e. g. [19]). This method exploits the fact that the summation over fermionic states in the vicinity of the Fermi surface separates into independent summations over energy and degeneracy variables. Since the relevant excitation energies are of the order of the CEF excitation \( \delta_{n}(H) \) the dominant contribution to the integral in Eq. (13) comes from electronic states with \( |\epsilon(\mathbf{p})| \sim \delta_{n}(0) \ll W \). First one replaces the integral over \( \mathbf{q} \) by an equivalent integration over \( \mathbf{p}’ = \mathbf{p} - \mathbf{q} \) where the external momentum \( \mathbf{p} \) is kept fixed. Then one restricts the frequency integration to a shell \( |\omega'| \ll \epsilon_{c} \) around the Fermi surface such that \( \delta_{n} \ll \epsilon_{c} \ll W \) is fulfilled. Here \( 2W \) is the conduction band width. In this shell we may approximate the momentum space integral by

\[
\int \frac{d^3p'}{(2\pi)^3} = \frac{N(0)}{4\pi} \int_{0}^{2\pi} d\theta' \int_{0}^{\pi} \sin \theta' d\theta' \int_{-\epsilon_{c}}^{\epsilon_{c}} d\epsilon' .
\]  

where \( \theta', \phi' \) are the polar and azimuthal angles of \( \mathbf{p}' \). Furthermore \( N(0) \) is the conduction electron density of states per spin at the Fermi energy (\( \epsilon_F = 0 \)). Using the analytical properties of the self-energy \( \Sigma(\omega) \) which imply that sign \( \text{Im} \Sigma(\omega) = -\text{sign} \omega \) the \( \epsilon' \)-integration gives for \( \delta_{n}(H) \)

\[
\Sigma(\omega) = g^2 N(0) \sum_{\alpha\beta,n} \langle \bar{q}_\alpha^2 \bar{q}_\beta^2 |\Gamma_5 |O_{\alpha\beta}| \Gamma_\pi^n \rangle^2 \int \frac{d\omega'}{2\pi} \frac{2\delta_n}{(\omega - \omega')^2 - \delta_n^2 + i\eta} \text{sign}(\omega') \arctan \frac{\epsilon_c}{\text{Im} \Sigma(\omega')} .
\]  

We solve the self-consistency equation for the imaginary part \( \text{Im} \Sigma(\omega) \)

\[
\text{Im} \Sigma(\omega) = -g^2 N(0) \sum_{\alpha\beta,n} \langle \bar{q}_\alpha^2 \bar{q}_\beta^2 |\Gamma_5 |O_{\alpha\beta}| \Gamma_\pi^n \rangle^2 \sum_{\rho = \pm 1} \text{sign}(\omega + \rho \delta_{n}(H)) \arctan \frac{\epsilon_c}{\text{Im} \Sigma(\omega + \rho \delta_{n}(H))} ,
\]  

from which we subsequently deduce the real part by Kramers-Kronig transformation. The explicit form immediately shows that

\[
| \text{Im} \Sigma(\omega) | \leq \pi g^2 N(0) \sum_{\alpha\beta,n} \langle \bar{q}_\alpha^2 \bar{q}_\beta^2 |\Gamma_5 |O_{\alpha\beta}| \Gamma_\pi^n \rangle^2 \frac{\hbar}{\tau} .
\]  

It is important to note that \( \text{Im} \Sigma(\omega) \) exhibits discontinuous jumps at the energies corresponding to the singlet-triplet excitations. This feature is a direct consequence of the assumption that the CEF excitations are long-lived and dispersionless bosonic excitations. Of particular interest is the discontinuity at \( \delta_{+}(H) \)

\[
| \text{Im} \Sigma(\delta_{+}(H) + \eta) - \text{Im} \Sigma(\delta_{+}(H) + \eta)| \geq \frac{\hbar}{\tau} \arctan \frac{\epsilon_c \tau}{\hbar} .
\]  

This discontinuity in the imaginary part inevitably implies a logarithmic singularity in the real part \( \text{Re} \Sigma(\omega) \) which, in turn, leads to an unphysical divergence in the effective mass for \( \delta_{+}(H) \rightarrow 0 \).

In the limit \( \epsilon_c \rightarrow \infty \) where \( 2 \arctan \frac{\epsilon_c}{\text{Im} \Sigma(\omega')} \rightarrow \pi \) the result agrees with that of non-selfconsistent second order perturbation theory. In this case differentiating the self energy with respect to \( \omega \) under the integral and using integration by parts one finally gets from Eq. (12):
The eigenstates are then given in Table I. The field dependence of \( \delta_n(H) \) is denoted by \( \delta_n(H) = E_n^+(H) - E_n^-(H) \). The eigenstates are given in terms of zero-field singlet-triplet states \( |0,0\rangle \) and \( |1,\pm\rangle, |1,0\rangle \) respectively (\( \hbar = g\mu_B H \)).

| eigenstate \( |\Gamma_n(H)\rangle \) | \( E(H) \) | \( \delta_n(H) \) |
|-----------------|-----------------|-----------------|
| \( |\Gamma^+_0(H)\rangle \) | \( u|0,0\rangle + v|1,0\rangle \) | \( \frac{1}{2}(\Delta - \tilde{\Delta}) \) | 0 |
| \( |\Gamma^+_1(H)\rangle \) | \( |1,\rangle \) | \( \Delta - h \), \( \frac{1}{2}(\Delta + \tilde{\Delta}) - h \) |
| \( |\Gamma^+_2(H)\rangle \) | \( u|1,0\rangle - v|0,0\rangle \) | \( \frac{1}{2}(\Delta + \tilde{\Delta}) \) | \( \Delta \) |
| \( |\Gamma^-_1(H)\rangle \) | \( |1,-\rangle \) | \( \Delta + h \), \( \frac{1}{2}(\Delta + \tilde{\Delta}) + h \) |

\[
m^* \frac{m}{m} = 1 + g^2 N(0) \bar{f}\chi Q(H);
\]

\[
\chi Q(H) = \sum_{\alpha\beta,n} 2 |\langle \Gamma_n(H)|O_{\alpha\beta}|\Gamma_n^+(H)\rangle|^2 \delta_n(H).
\] (21)

The directional average (with respect to polar and azimuthal angles \( \theta, \phi \) of \( \hat{q} \)) for quadrupolar form factors \( \bar{f} = \langle \tilde{q}_0^2 \tilde{q}_0^2 \rangle = \frac{1}{15} \) is a constant. Furthermore \( \chi Q(H) \) in Eq. (21) is the field-dependent static uniform quadrupolar susceptibility. Note that the form factor average can be trivially factored out as a constant (1/15) only because in the present local approximation the boson propagator is momentum independent, i.e. the CEF excitations are dispersionless. The more general case will be discussed below.

IV. FIELD DEPENDENCE OF THE EFFECTIVE MASS: DISPERSIONLESS MODEL

When a magnetic field is applied to the sample the field dependence of the effective mass is completely determined by that of the quadrupolar susceptibility in Eq. (21). To calculate this quantity one first has to know the singlet-triplet excitation energies \( \delta_n(H) \) and the eigenstates and matrix elements in applied field. They were given by Shiina et al. in closed form for field applied along cubic symmetry directions. We use these results in the following. The CEF eigenstates are denoted as singlet \( |\Gamma^+_s\rangle \) and triplet \( |\Gamma^+_t\rangle \) \((n = +,0,-)\), respectively. The CEF and Zeeman Hamiltonian can be easily mapped to a pseudospin basis \( |\bar{u},\bar{v}\rangle \) and then diagonalised. In pseudospin basis the zero-field singlet is denoted by \( |0,0\rangle \) and the triplet by \( |1,m\rangle \) \((m=1,0,-1)\). For field \( H \parallel [001] \) the field-split CEF levels and mixed eigenstates are then given in Table I. The field dependence of \( \delta_n(H) \) has recently been determined by INS experiments.

The mixing coefficients \( u, v \) are determined by the matrix elements of the dipolar operator \( J \) in the Zeeman term which may be expressed by \( \alpha = 5/2 - 2d^2, \beta = 2\sqrt{5/3}d, \delta = \beta/\alpha \). They are given by

\[
v = -\text{sgn}(y)[\frac{1}{2}(1 - \frac{\Delta}{\tilde{\Delta}})]^{1/2}; \quad u = (1 - v^2)^{1/2}
\]

\[
\Delta = [\Delta^2 + 4\delta^2h^2]^{1/2}; \quad h \equiv g\mu_B H
\] (22)

Note that a finite mixing \( v \neq 0 \) occurs only due to the tetrahedral CEF contribution \( (y \neq 0) \) which leads to \( d \neq 0 \) in Eq. (2) and hence \( \delta \neq 0 \). When \( d = 0 \) \( (\delta = 0) \) there is no mixing between \( |0,0\rangle \) and \( |1,0\rangle \) and consequently the energies of \( |\Gamma^+_s\rangle \) and \( |\Gamma^+_t\rangle \) will be independent of the field \( H \). For nonzero \( d \) and \( v \) these two levels will repel with increasing field \( H \). The other two triplet states \( |\Gamma^+_s\rangle \) have a linear Zeeman splitting of \( 2h \) independent of \( d \). For \( \delta \geq 0 \) the singlet ground state level \( E_s \) and lowest triplet level \( E_t^+ \) cross at a critical field \( h_c = \Delta/(1 - \delta^2) \) meaning \( \delta_n(h_c) = 0 \).

For evaluation of the effective mass we need the quadrupolar matrix elements in Eq. (21). They may all be expressed in terms of the irreducible zero field matrix elements \( \alpha' = \sqrt{\Delta^2(13 - 20d^2)} \) and \( \beta' = \sqrt{35(1 - d^2)} \). With their help and defining \( \delta' = \alpha'/\beta' \) one obtains the following nonzero matrix elements:

\[
|\langle \Gamma_s|O_{yz}|\Gamma^+_t\rangle|^2 = |\langle \Gamma_s|O_{zx}|\Gamma^-_t\rangle|^2 = \frac{1}{2}\beta^2(u - \delta'v)^2 = |m_Q^-|^2
\]

\[
|\langle \Gamma_s|O_{yx}|\Gamma^-_t\rangle|^2 = |\langle \Gamma_s|O_{zx}|\Gamma^+_t\rangle|^2 = \frac{1}{2}\beta^2(u + \delta'v)^2 = |m_Q^0|^2
\]

\[
|\langle \Gamma_s|O_{xy}|\Gamma^+_t\rangle|^2 = \beta^2 = |m_Q^+|^2
\] (23)
FIG. 3: Left (a): Dispersion of lowest triplet quadrupolar exciton $\omega_{\uparrow}(q)$ at the critical field $h_c(g_Q)$ where $\omega_{\uparrow}(Q)$ becomes soft ($Q$ in units of $2\pi/a$). As $g_Q$ is reduced the dispersion becomes flat increasing the phase space for low energy conduction electron scattering. Right (b): Field dependence of $\Delta \chi_Q \sim \delta m^*/m$ for various strengths of intersite quadrupolar coupling $g_Q$ (left). For small $g_Q$ mass renormalisation close to $h_c$ is large due to flat $\omega_{\uparrow}(q)$ dispersion. For larger $g_Q$ the dispersion becomes stronger and $\omega_{\uparrow}(q)$ softens only in the vicinity of $Q = (0,0,1)$ leading to a much smaller mass enhancement at $h_c$. The curves correspond to $g_Q$ given in the legend in decreasing order. For the value $g_Q = 0.3$ corresponding to PrOs$_4$Sb$_{12}$ little field dependence of $\delta m^*/m$ remains.

Inserting the matrix elements and excitation energies in Eq. (21) and using $|m_Q^+|^2 + |m_Q^-|^2 = \beta^2[1 - v^2(1 - \delta^2)]$ we finally obtain the expression

$$\chi_Q(H) = \frac{2\beta^2}{\Delta} \left[ \frac{2\Delta\Delta'}{\Delta^2 - h^2}[1 - v^2(1 - \delta'^2)] + \frac{\Delta}{\Delta} \right]$$

(24)

where we defined $\Delta' = \frac{1}{2}(\Delta + \tilde{\Delta})$. Without tetragonal CEF ($d^2 = 0$) we have $\Delta' = \tilde{\Delta} = \Delta$ and then the above expression reduces to

$$\chi_Q(H) = \frac{2\beta^2}{\Delta} \left[ \frac{2\Delta^2}{\Delta^2 - h^2}[1 - v^2(1 - \delta'^2)] + 1 \right]$$

(25)

For small fields ($h \ll \Delta$) the general $\chi_Q(H)$ in Eq. (25) may be expanded with a leading term $\sim (h/\Delta)^2$ according to

$$\chi_Q(H) \approx \frac{2\beta^2}{\Delta} \left\{ 3 + [2\delta^2(\delta'^2 - 3)] \left( \frac{h}{\Delta} \right)^2 \right\}$$

(26)

The zero field mass enhancement without tetragonal CEF ($d^2 = 0$) is then obtained from Eq. (21,25) simply as

$$\frac{m^*}{m} = 1 + g^2 N(0) 3f \beta^2 \frac{2\beta^2}{\Delta}$$

(27)

The states and energies in Table I are nominally derived for $H \parallel [001]$. However it was shown in Refs. 3, 11 that for low fields ($h < \Delta$) they are the same for all field directions, i.e. approximately isotropic. Therefore the quadrupolar susceptibility derived above and the related mass enhancement are also approximately isotropic as long as $h$ is appreciably below $\Delta$. This condition is required anyway in the dispersionless case where the calculation is only valid for moderate fields when $\delta_{\uparrow}(h)$ is still large enough.

V. INFLUENCE OF QUADRUPOLE EXCITON DISPERSION ON THE MASS ENHANCEMENT

In the previous section we investigated a model of noninteracting local singlet-triplet quadrupole excitations. For appreciably large concentration of Pr ions in the system Pr$_{1-x}$La$_x$Os$_4$Sb$_{12}$ this is no longer justified. Due to effective
interactions between the 4f states on different sites the singlet-triplet excitations at \( \Delta \) acquire a dispersion. Formally this is already included in the self energy of Eq. (13) provided the boson propagator for a dispersive mode is used by replacing \( \delta_n \rightarrow \omega_n(q) \) in Eq. (15). The dispersion is due to quadrupolar RKKY-type intersite interactions which are obtained in second order perturbation theory from \( H_{AC} \) and given by \[ H_Q = \sum_{\langle ij \rangle} K_Q(ij)O(i) \cdot O(j) \] (28)

where \( O = (O_{yz}, O_{zx}, O_{xy}) \) is the \( \Gamma_5 \) type quadrupole. The sum is restricted to nearest neighbors and \( K_Q \) is the effective quadrupolar coupling constant. This interaction leads to the field induced antiferroquadrupolar order from which the \( \Gamma_5 \) symmetry has been inferred [9]. Formally \( H_Q \) may be obtained from an RKKY type mechanism in order \( \sim g^2 \) in the coupling constant of \( H_{AC} \). In practice the n.n. term \( K_Q \) is determined from the experimentally observed dispersion of the quadrupolar excitons \( \omega_n(q) \) which are fully degenerate for zero field [21]. In finite field the dispersive excitation branches are obtained by replacing the Hamiltonian in Eq. (9) with \( H_{CEF} + H_Q + H_Z \). Using a generalised Holstein-Primakoff approximation [12] the three quadrupolar exciton modes at moderate fields are described by

\[
\omega_{\pm}(q) = \omega(q) \mp h; \quad \omega_0(q) = \omega(q)
\] (29)

where the zero-field dispersion is given by

\[
\omega(q) = \sqrt{A_q^2 - B_q^2} \\
A_q = \Delta + zK_Q\gamma_q; \quad B_q = -zK_Q\gamma_q \\
\gamma_q = \cos \frac{1}{2} q_x \cos \frac{1}{2} q_y \cos \frac{1}{2} q_z
\] (30)

Here \( z=8 \) is the coordination number and \( \gamma_q \) the structure function of the bcc cubic lattice of Pr ions with momentum \( q \) measured in r.l.u. \( (2\pi/a) \). The width of the exciton dispersion is controled by the effective quadrupolar coupling constant \( K_Q \) or in dimensionless form by \( g_Q = z\beta^2K_Q/\Delta \). From the analysis of the AFQ phase diagram [12] and experimental zero-field dispersion [21] one may deduce \( g_Q \approx 0.3 \) in \( \text{PrOs}_4\text{Sb}_{12} \). The minimum of the dispersion occurs at the bcc zone boundary wave vector \( Q = (1,0,0) \) (r.l.u.). The zero-field energy is given by \( \omega(Q) = \Delta[1 - 2g_Q]^2 \). Consequently the soft mode indicating transition to (zero-field) AFQ order would occur at \( g_Q = 0.5 \) which is larger than the above value of 0.3 for pure \( \text{PrOs}_4\text{Sb}_{12} \). Therefore application of a magnetic field is necessary to achieve a soft mode \( \omega_{\pm}(Q) = 0 \) at a critical field \( h_c \). The dispersions in Eq. (31) are approximations where the field dependence of the \( \Gamma_x - \Gamma_7 \) splitting has been neglected. This is possible as long as their dipolar
matrix element $d^2, \delta^2 \ll 1$ which is true for the case $d^2 = 0.067$ (Fig. 2 (left)). Then the soft mode condition leads to the approximate critical field $h_c/\Delta = (1 - 2g_Q)^\frac{1}{2}$ above which AFQ order will be induced. Using $g_Q = 0.3$ leads to $h_c/\Delta = 0.586$ which is close to the exact value 0.632 given in Ref. [12].

Calculation of electron self energy and mass enhancement in the dispersive case proceeds now exactly along the lines described in Sect. [11]. The main modifications arise from the fact that a more sophisticated approximation for the electron-quadrupolar exciton spectral function is employed. For large cut-off energies $\epsilon_c \to \infty$ one obtains

$$\frac{m^*}{m} = 1 + g^2 N(0) \sum_{\alpha\beta,n} 2\langle |D_{\alpha\beta}| |\Gamma^\alpha_n| \rangle^2 \frac{1}{2\pi} \int d\Omega_q \frac{\hat{q}_x^2 \hat{q}_y^2}{\omega_n(q)}$$

which closely parallels the expression derived by Nakajima and Watabe [22] for the effective mass enhancement due to electron-phonon interaction. Here we use $q = \hat{q} q$ with $q = 2p_F \cos \theta$ (Fig. 1) where $\hat{q}$ has polar and azimuthal angles $\theta$ and $\phi$, respectively. Due to the geometric restrictions only half the solid angle $(2\pi)$ contributes in the momentum integral. Replacing the dispersive modes $\omega_n(q)$ by the dispersionless singlet-triplet excitation energies $\delta_n$ leads to the previous result in Eqs. (21,25). Using the explicit matrix elements and dispersions we may again represent the mass enhancement in the form of Eq. (21).

$$\frac{m^*}{m} = 1 + g^2 N(0) \int \chi_Q(H)$$

$$\chi_Q(H) = 2\beta^2 \frac{1}{\Delta} \left\{ [1 - v^2(1 - \delta^2)] \frac{1}{2\pi} \int d\Omega_q \frac{\Delta \omega(q)}{\omega(q)^2 - h^2 (\hat{q}_x^2 + \hat{q}_y^2) + \frac{1}{2\pi} \int d\Omega_q \frac{\Delta \omega(q)}{\omega(q)^2 - h^2 (\hat{q}_x^2 + \hat{q}_y^2)} \right\}$$

Here the first and second terms are due to the virtual excitations of $\omega_\pm(q)$ and $\omega_0(q)$ bosons respectively. In the dispersionless limit this reduces to the previous result in Eq. (25) for the case $d^2 = 0$ (no tetragonal CEF) which corresponds to the present treatment due to the neglect of the $E_s, E_t^0$ level repulsion implied in the dispersions of Eq. (29). The above expression for the mass enhancement have to be evaluated numerically due to the BZ integrations. This will be discussed in the next section. But we may nevertheless gain some qualitative insights by simple approximations to these integrals in the zero-field case. For that purpose we expand $\omega(q)$ around one of the six equivalent zone boundary X- points with $Q = (\pm 1, 0, 0)$ etc.. Then one obtains an isotropic approximate dispersion given by

$$\omega(q')^2 = \omega_Q^2 + \omega_0^2 (\pi q')^2$$

where $q' = Q - q$ is the momentum vector counted from the X-point and $q'$ is its length. Furthermore $\omega_Q = \Delta [1 - 2g_Q]^{\frac{1}{2}} = h_c$ and $\omega_0 = \sqrt{\Delta Q C}$. On approaching the critical $g_Q = \frac{1}{2}$ the soft mode frequency $\omega_Q$ vanishes. In this limit the integral in Eq. (32) may easily be evaluated. The approximate Fermi surface geometry of PrOs$_4$Sb$_{12}$ shows that $p_F \approx \frac{1}{\sqrt{2}}$ (r.l.u.) or $2p_F = \sqrt{2}$ (Fig. 1). Therefore $2p_F > Q$ which means that the minimum in $\omega(q)$ is included in the domain of the momentum integrals in Eqs. (32,33). We restrict the latter to the sphere around the minimum at $Q$ (X-point) with a cutoff radius given by $q_c^* < 1$ where only $1/2$ contributes due to geometric restrictions. We then obtain from Eq. (32)

$$\frac{m^*}{m} \approx 1 + g^2 N(0) \left( \frac{q_c^*}{\omega_0} \right)^4 \frac{2\beta^2}{\pi}$$

where the momentum cutoff $q_c^*$ around X is defined such that the quadratic expansion in Eq. (34) is still valid. Note that although the exciton energy becomes soft $\omega_Q \to 0$ at the zone boundary there is no divergence in the mass renormalisation. This is due to the small phase space volume around the X-point which gives only a small contribution despite the vanishing exciton frequency. In addition the singular contribution is suppressed by the fact that the form factor $\hat{q}_x^2 \hat{q}_y^2$ vanishes exactly at the X-point directions and only contributions from its environment are picked up by the integration. The above expression is formally quite similar to the dispersionless result of Eq. (27). In the latter case the renormalisation diverges when $\Delta \to 0$ because this corresponds to a softening in the whole BZ. Thus we conclude from Eqs. (27,34) that the inclusion of a mode dispersion removes the problem of singular mass renormalisation, $m^*/m$ stays finite for all coupling constants $g_Q$, even when $g_Q = g_Q^* = 0$ when the exciton frequency becomes soft at $Q$. However the above formula cannot give a reliable estimate for $m^*/m$ due to the strong momentum cutoff dependence, we therefore have to employ a numerical evaluation of Eq. (35).
because the softening becomes strongly constricted around and the mass enhancement is finite, though still large at
becomes soft for all \( q \). This corresponds to the right magnitude for thermal mass enhancement.

level repulsion of \( \Gamma_s \) matrix elements leads to a field dependence of \( \chi \) enhancement mechanism. Using
introduced the dimensionless quadrupolar coupling constant \( g \equiv \frac{Q}{h} \). For larger \( d \), the level repulsion due to the tetrahedral CEF
strongly increases. For \( \text{PrOs}_4\text{Sb}_12 \) (\( g_d = 0.3 \)) one may expect little field dependence of \( \delta m^*/m \) between \( h = 0 \) and \( h = h_c \). The inset shows the dependence of the AFQ critical field \( h_c \) on \( g_Q \) with the approximation \( d^2 \approx 0 \).

VI. NUMERICAL RESULTS AND DISCUSSION

We first discuss the mass enhancement in the large band width approximation \( \epsilon_c \rightarrow \infty \) for dispersionless undamped CEF excitations. The absolute value of \( m^*(h)/m = 1 + \delta m^*/m \) is determined by the effective coupling constant \( \tilde{g} = g^2 N(0) \). Approximating \( N(0) \approx 1/2W \) (\( 2W \)=band width) this may be written as \( \tilde{g}/\Delta = \lambda^2(2W/\Delta) \). Here we introduced the dimensionless quadrupolar coupling constant \( \lambda = \frac{g}{\sqrt{W}} \). Assuming typical values of \( W = 1 \) eV, \( \lambda \approx 0.02 \) and using \( \Delta = 8 \) K we obtain \( \tilde{g}/\Delta \tilde{f} \approx 0.077 \) as the size of the effective coupling for the quadrupolar mass enhancement mechanism. Using \( d^2 = 0.067 \) and hence \( \beta^2 \approx 32.6 \) we obtain a zero field enhancement of \( m^*/m \approx 16 \).

This corresponds to the right magnitude for thermal mass enhancement.

For discussion of the field dependence we use the quadrupolar susceptibility which contains only \( d \) as adjustable parameter to avoid specifying \( \tilde{g} \). In the case of weak tetrahedral CEF such as realised in \( \text{Pr}_{1-x}\text{La}_x\text{Os}_4\text{Sb}_12 \) the level repulsion of \( \Gamma_s \) and \( \Gamma_0 \) is also weak and therefore the \( \Gamma^+_t \) level crosses the \( \Gamma_s \) ground state at a critical field \( h_c = \frac{\Delta}{1 - \beta^2} \) in the dispersionless case. The decrease in the excitation gap for \( h < h_c \) and the field dependence of matrix elements leads to a field dependence of \( \chi(q) \) (Eq. 21) which is shown in Fig. 2 for \( d^2 = 0 \) and \( d^2 = 0.067 \). For larger \( d^2 \) the increase is diminished and eventually for \( d^2 > 0.42 \) the level repulsion due to the tetrahedral CEF is strong enough to lead to an increase in excitation energy and hence to a decreasing effective mass. Likewise the level repulsion prevents field induced AFQ order, therefore it is not appropriate for concentrated \( \text{PrOs}_4\text{Sb}_12 \). This case resembles more that of pure metallic \( \text{Pr} \) where a singlet-singlet level repulsion in a field also leads to a decrease in quasiparticle mass \[13\].

Let us next turn to the divergent mass renormalization which is predicted for dispersionless undamped CEF excitations when the triplet level \( E^+_t \) approaches the singlet ground state level \( E_s \) (Fig. 2). Therefore in the (level crossing) case \( d^2 < 0.42 \) this approach is only valid for moderate fields. As the divergence follows directly from the general analytic structure of the corresponding electron self energy it persists also in the self consistent solution for finite band width. Selfconsistency leads to an overall reduction of the mass renormalization as can be seen from Fig. 4.

The (unphysical) divergence of the mass enhancement close to the critical magnetic field is an artefact of the model which assumes dispersionless undamped CEF excitations. Inelastic neutron scattering [21] however, have shown that the singlet-triplet excitations have a pronounced dispersion. The dispersive width corresponds to \( \sim 40\% \) of the CEF excitation energy \( \Delta \). An applied field of critical strength therefore leads to a softening of \( \omega(q) \) only in the restricted phase space around the AFQ ordering vector \( Q \). Consequently the mass renormalisation will be finite even at the critical field \( h_c \) for AFQ order when \( \omega(Q) = 0 \). This is shown in Fig. 3 for various effective quadrupolar coupling strengths \( g_Q \). For \( g_Q = 0 \) the mass renormalisation at \( h_c \) would diverge as in Fig. 2 because the excitation energy becomes soft for all \( q \)-vectors. For small \( g_Q \) and dispersive width the softening appears only around \( Q \) (Fig. 3) and the mass enhancement is finite, though still large at \( h_c \). It is progressively diminished with further increasing \( g_Q \) because the softening becomes strongly constricted around \( Q \) (see Fig. 3b). For the value \( g_Q = 0.3 \) appropriate for
PrOs\textsubscript{4}Sb\textsubscript{12} the dispersion is sufficiently large to suppress the field dependence of $\delta m^*/m$ as depicted in Fig. 8h (full line).

An alternative presentation of results for the dispersive case is shown in Fig. 5. The mass enhancement is shown as function of quadrupolar coupling $g_Q$ ($\sim$ dispersive width) for the two limiting cases $h = 0$ and $h = h_c(g_Q)$. For large $g_Q$ and dispersion the field variation of $\delta m^*/m$ between $h = 0$ and $h = h_c$ becomes small. This is partly due to the fact that $h_c(g_Q)$ itself becomes small for large $g_Q$ (see inset of Fig. 5). When $g_Q$ decreases the difference in $\delta m^*/m$ for $h = 0$, $h_c$ increases rapidly because the mass enhancement at $h = h_c$ becomes singular when approaching the dispersionless case $g_Q \rightarrow 0$. The arrow corresponds to the proper value of $g_Q$ for PrOs\textsubscript{4}Sb\textsubscript{12} and it shows again that one should expect little field dependence of the mass enhancement in this case.

VII. CONCLUSION AND OUTLOOK

In this work we have studied in detail the quasiparticle mass enhancement originating in the aspherical Coulomb scattering of conduction electrons from singlet triplet CEF excitations. This model has some relevance for the heavy fermion superconductor PrOs\textsubscript{4}Sb\textsubscript{12} where the Pr$^{3+}$ 4f states are subject to a tetrahedral CEF leading to a singlet ground state and an excited triplet. For small tetrahedral CEF the latter has a mostly nonmagnetic character and therefore may be excited by aspherical Coulomb scattering from conduction electrons. These virtual second order processes lead to a quasi particle mass renormalisation which may well be the source of the large thermal and dHvA effective masses observed in PrOs\textsubscript{4}Sb\textsubscript{12}. A hybridisation mechanism between conduction and 4f electrons can be ruled out since the Fermi surface of PrOs\textsubscript{4}Sb\textsubscript{12} is identical to that of LaOs\textsubscript{4}Sb\textsubscript{12} which advocates for fully localised 4f electrons in Pr. Indeed the well defined CEF excitations seen in INS \cite{21} support this view.

If aspherical Coulomb scattering of conduction electrons plays a role in the mass enhancement one should expect a field dependence of the latter. For small enough tetrahedral CEF characterised by the parameter $d^2 \ll 1$ the lowest triplet component crosses the singlet ground state at a critical field $h_c$. The mass enhancement in second order perturbation theory with respect to $H_{AC}$ then increases with field and becomes singular at $h_c$. For larger tetrahedral CEF ($d^2 > 0.42$) the excitation energy between singlet ground state increases with field leading to a decrease of the mass enhancement, similar as has been observed in Pr metal where the mass renormalisation is due to exchange scattering from a singlet-singlet CEF level scheme.

The observed singular mass enhancement close to the critical field of level crossing is an artefact of the dispersionless model, both in the perturbative and selfconsistent treatment. Any dispersion of the singlet-triplet excitations due to effective intersite quadrupolar interactions will lead to a finite effective quasiparticle mass. We have shown that the enhancement decreases strongly with increasing dispersion because the phase space for conduction electron scattering from low lying CEF excitations (quadrupolar excitons) is constrained to the wave vector $Q$ of incipient field induced AFQ order. For a quadrupolar coupling constant $g_Q = 0.3$ corresponding to PrOs\textsubscript{4}Sb\textsubscript{12} the field dependence is reduced to a few percent. In addition this compound is superconducting below $T_c = 1.85$ K with $H_{c2} = 2.2$ T and has a field induced AFQ phase above $H_{c} = 4.5$ T. Therefore only a reduced field range is left to observe the small field dependence possible at $g_Q = 0.3$. We conclude that the concentrated PrOs\textsubscript{4}Sb\textsubscript{12} is not a favorable system to observe the field dependent mass enhancement due to aspherical Coulomb scattering.

A more promising system may be the La-diluted systems Pr$_{1-x}$La$_x$Os\textsubscript{4}Sb\textsubscript{12}. On increasing $x$ the average distance between the Pr 4f shells becomes larger and therefore the effective quadrupolar coupling $g_Q(x)$ will decrease with $x$, i.e. the dispersion of the 4f CEF excitons will progressively decrease. This means that the field dependence of effective masses will become more pronounced according to Figs. 8a,8b. Of course the absolute (zero-field) size of the mass enhancement is also reduced since the self energy in Eq. 17 will be proportional to the number $(1-x)$ of Pr sites. There should however be an intermediate concentration region for $x$ where the field dependence is pronounced ($g_Q$ small) and the $\gamma(x)$ still large enough as compared to the other (lattice or CEF-Schottky) contributions such that the field dependence of $\gamma(x,H)$ is experimentally accessible. Furthermore in this region of $x$ one may probe a larger field range because there is no more AFQ order present. Therefore we propose that the field dependence of the electronic specific heat in mixed crystals of Pr$_{1-x}$La$_x$Os\textsubscript{4}Sb\textsubscript{12} is systematically investigated and analysed. It may hold important clues to the microscopic origin of the large effective mass in the concentrated compound PrOs\textsubscript{4}Sb\textsubscript{12}. 

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