Field-dependent mass enhancement in $Pr_{1-x}La_xOs_4Sb_{12}$ 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 well-known 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 $Pr_{1-x}La_xOs_4Sb_{12}$ where close to $x=1$ the compound develops heavy quasiparticles with a large specific heat γ. There the low-lying CEF states are a 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 $Pr_{1-x}La_xOs_4Sb_{12}$ with suitably large x when the dispersion is small.

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I. INTRODUCTION

The filled-skutterudite compound $PrOs_4Sb_{12}$ has recently obtained considerable attention. There are several reasons for that. It is a heavy fermion ($\gamma\sim350-500$ mJ/mol K^2) superconductor with a transition temperature of $T_c(\mathrm{Pr})=1.85$ K. This temperature is larger than that of the related system $LaOs_4Sb_{12}$ which is $T_c(\mathrm{La})=0.74$ K. A number of experiments, such as those on Sb-NMR relaxation rate in $Pr_{1-x}La_xOs_4Sb_{12}$,[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 suggests a possible nodal structure while the latter in addition points toward two distinct superconducting phases. The observed enhancement of the superconducting transition temperature by more than a factor of 2 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 such as $LaPb_3$, it suppresses the superconducting transition temperature rather efficiently. So why does it enhance T_c in the present case? Since the phonons in $LaOs_4Sb_{12}$ and $PrOs_4Sb_{12}$ are very nearly the same, the enhancement must come from the two $4f$ electrons which Pr^{3+} has. The heavy fermion behavior of $PrOs_4Sb_{12}$ seems puzzling too. It shows up in a large specific-heat jump $\Delta C/T_c\simeq500$ mJ/(mol K^2) at T_c and also in a large effective mass in de Haas–van Alphen (dHvA) experiments. The Kondo effect cannot be the origin of the heavy quasiparticles since the $4f^2$ electrons are well localized with 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 quasiparticles excitations lies in the crystalline electric field (CEF) splitting of the $J=4$ multiplet which will be discussed in Sec. II, together with the 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 Sec. 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 Sec. IV for dispersionless excitations and in Sec. V for the case with dispersive quadrupolar excitons. Some numerical results are discussed in Sec. VI, and Sec. VII finally gives the conclusions.

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

From inelastic neutron scattering (INS) 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,

$$|\Gamma_1\rangle=\frac{\sqrt{30}}{12}(|+4\rangle+|-4\rangle)+\frac{\sqrt{21}}{6}|0\rangle, \tag{1}$$

with a low-lying triplet excited state at an energy of $\Delta=8$ K.[6,8–10] The other CEF levels are so high in energy that they can be neglected. The Γ_t triplet state of T_h symmetry is a superposition of two triplets Γ_4 and Γ_5 of O_h symmetry. More specifically one finds[9,10]

$$|\Gamma_t,m\rangle=\sqrt{1-d^2}|\Gamma_5,m\rangle+d|\Gamma_4,m\rangle, \quad m=1,\ldots,3, \tag{2}$$

with states of O_h symmetry given by

$$|\Gamma_5,\pm\rangle=\pm\sqrt{\frac{7}{8}}|\pm3\rangle\mp\sqrt{\frac{1}{8}}|\mp1\rangle,$$

$$|\Gamma_5,0\rangle=\sqrt{\frac{1}{2}}(|+2\rangle-|-2\rangle),$$

$$|\Gamma_4,\pm\rangle = \mp\sqrt{\frac{1}{8}}|\mp 3\rangle \mp \sqrt{\frac{7}{8}}|\pm 1\rangle, \tag{3}$$

$$|\Gamma_4,0\rangle = \sqrt{\frac{1}{2}}(|+4\rangle - |-4\rangle).$$

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

$$H_{\mathrm{ex}}(i) = -2(g_J-1)J_{\mathrm{ex}} \sum_{\mathbf{kq}\sigma\sigma'} (\mathbf{s}_{\sigma\sigma'}\cdot\mathbf{J}_i)c^{\dagger}_{\mathbf{k}-q\sigma'}c_{\mathbf{k}\sigma} \tag{4}$$

where $c^{\dagger}_{\mathbf{k}\sigma}(c_{\mathbf{k}\sigma})$ are the creation (annihilation) operators for conduction electron with momentum $\mathbf{k}$ and spin σ while $\mathbf{s}$ is their spin operator. Furthermore $\mathbf{J}$ is the Pr total angular momentum (J=4) and g_J is the Landé factor. The aspherical Coulomb interaction in local-orbital basis is given by[11]

$$H_{\mathrm{AC}}(i) = \left(\frac{5}{4\pi}\right)^{1/2} \sum_{kk'\sigma}\sum_{m=-2}^{+2} I_2(k's;kd) \times Q_2[Y_2^m(\mathbf{J}_i)c^{\dagger}_{k's\sigma}c_{kdm\sigma} + \mathrm{H.c.}]. \tag{5}$$

Here $c_{klm\sigma}$ destroys a conduction electron with momentum $k=|\mathbf{k}|$ in a $l=2$ state with azimuthal quantum number m and spin σ, and $c^{\dagger}_{k's\sigma}$ 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 $Y_2^m(\mathbf{J})$ are given by

$$Y_2^0 = (2/3)^{1/2}[3J_z^2 - J(J+1)]/N_J,$$

$$Y_2^{\pm} = \pm(J_zJ^{\pm} + J^{\pm}J_z)/N_J,$$

$$Y_2^{\pm 2} = (J^{\pm})^2/N_J, \tag{6}$$

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 $PrOs_4Sb_{12}$ 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.[12] With this information the two features pointed out above, i.e., the increase in T_c when La is replaced by Pr and the heavy quasiparticle mass, can be understood quantitatively.[7] 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 in the $4f^2$ wave function.

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 $Pr_{1-x}La_xOs_4Sb_{12}$ the aspherical 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 paper 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 in the excitation energy of one of the three states, at least for small tetrahedral admixture d as in $PrOs_4Sb_{12}$. Therefore an increase in 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 realized for $|d|>0.65$.

III. SELF-ENERGY AND MASS RENORMALIZATION

We start out by specifying the electronic part of the Hamiltonian for the system $La_{1-x}Pr_xOs_4Sb_{12}$. It is of the type

$$H = H_{\mathrm{el}} + H_{\mathrm{CEF}} + H_{\mathrm{AC}} + H_{\mathrm{ex}} + H_Z. \tag{7}$$

Here H_{el} is of the conventional form and need not be explicitly written down. It contains the conduction-band dispersion which may be described by a next-nearest-neighbor (NNN) tight-binding model[15] according to

$$\epsilon_{\mathbf{k}\sigma} = t\cos\frac{1}{2}k_x\cos\frac{1}{2}k_y\cos\frac{1}{2}k_z + t'(\cos k_x + \cos k_y + \cos k_z), \tag{8}$$

with $t=174$ meV and $t'=-27.84$ meV. The transfer integrals t and t' are chosen so as to reproduce the observed linear specific-heat coefficient $\gamma=36$ mJ/(mol K^2) of the non-f reference compound $LaOs_{12}Sb_{12}$.[16] In the *electron* picture the associated Fermi surface (FS) consists of H-centered spheroids with a Fermi radius $p_F\simeq 0.7(2\pi/a)$, see Fig. 1(b). Aside from subtle effects this is quite similar to the local-density approximation (LDA) FS in Ref. 17 [note that in this reference the FS is depicted in the *hole* picture as in Fig. 1(a)]. It corresponds to a single band originating in Sb $4p$ states.

The CEF and Zeeman Hamiltonians are

$$H_{\mathrm{CEF}} + H_Z = \sum_{i,\Gamma n} E_{\Gamma}|\Gamma_n(i)\rangle\langle\Gamma_n(i)| + g_J\mu_B\sum_i \mathbf{J}(i)\cdot\mathbf{H}. \tag{9}$$

The external magnetic field is denoted by $\mathbf{H}$, g_J is the Landé factor, and μ_B is the Bohr magneton. Furthermore i labels the Pr sites and $|\Gamma_n\rangle$ denotes the singlet ground state $|\Gamma_s\rangle$ and the triplet $|\Gamma_t^n\rangle$ [see Eq. (2)] with energies $E_s=0$ and $E_t=8$ 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

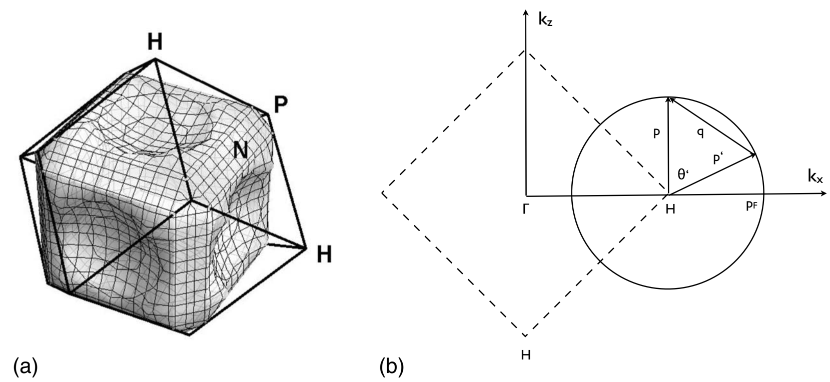

FIG. 1. Left (a): Fermi surface of NNN tight-binding model in hole representation in the bcc Brillouin zone (BZ). Right (b): schematic Fermi surface in electron representation in the two-dimensional (2D) projected Brillouin zone. It consists of spheroids around the equivalent H points $(\frac{2\pi}{a},0,0)$.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

states of cubic symmetry [see Eqs. (2) and (3)] we specialize Eq. (5) to cubic symmetry. In that case the aspherical Coulomb interaction written in a basis of Bloch states becomes

$$H_{\mathrm{AC}}(i)=g\sum_{\mathbf{k}q\sigma}\sum_{\substack{\alpha\beta\,cycl.\\ \sigma}}O^{i}_{\alpha\beta}\hat{q}_{\alpha}\hat{q}_{\beta}c^{\dagger}_{\mathbf{k}-q\sigma}c_{\mathbf{k}\sigma}e^{i\mathbf{k}\mathbf{R}_i},\qquad(10)$$

with $O_{\alpha\beta}=\frac{\sqrt{3}}{2}(J_\alpha J_\beta+J_\beta J_\alpha)$, $\hat{q}_\alpha=q_\alpha/|\mathbf{q}|$, and $\alpha\beta=yz,zx,xy$ denoting the three quadrupole operators with Γ_5 symmetry. The remaining Γ_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 Eq. (10) 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 the conduction electrons,

$$G(\mathbf{p},\omega)=\frac{1}{\omega-\epsilon(\mathbf{p})-\Sigma(\mathbf{p},\omega)}.\qquad(11)$$

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

$$\frac{m^*}{m}=1-\left.\frac{\partial\Sigma(p_F,\omega)}{\partial\omega}\right|_{\omega=0}\qquad(12)$$

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 $\Sigma(\mathbf{p},\omega)$ due to H_{AC} is given by

$$\Sigma(\mathbf{p},\omega)=g^2\sum_{\alpha\beta,n}\int\frac{d^3q}{(2\pi)^3}|\Lambda^n_{\alpha\beta}(\hat{\mathbf{q}})|^2\int\frac{d\omega'}{2\pi}D_n(\mathbf{q},\omega)G(\mathbf{p}-\mathbf{q},\omega-\omega').\qquad(13)$$

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 Ruderman-Kittel-Kasuya-Yoshida (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 $\Lambda^n_{\alpha\beta}(\hat{\mathbf{q}})$ is due to the quadrupolar Γ_5 form factors in Eq. (10). It is defined as

$$\Lambda^n_{\alpha\beta}(\hat{\mathbf{q}})=g\hat{q}_\alpha\hat{q}_\beta\langle\Gamma_s|O_{\alpha\beta}|\Gamma^n_t\rangle.\qquad(14)$$

The self-energy due to exchange scattering which involves the magnetic susceptibility can be safely neglected because of the smallness of d^2 [see Eq. (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

$$D_n(\mathbf{q},\omega)=D_n(\omega)=\frac{2\delta_n}{\delta_n^2-\omega^2}.\qquad(15)$$

Here the field-dependent singlet-triplet excitation energies are given by $\delta_n(H)=\epsilon^n_t(H)-\epsilon_s(H)$ $(n=+,0,-)$.

The self-energy in Eq. (13) can be evaluated following Migdal's integration procedure (see, e.g., Ref. 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 $\delta_\alpha\ll\epsilon_c\ll W$ is fulfilled. Here $2W$ is the conduction bandwidth. In this shell we may approximate the momentum space integral by

$$\int\frac{d^3p'}{(2\pi)^3}=\frac{N(0)}{4\pi}\int_0^{2\pi}d\phi'\int_0^{\pi}\sin\theta' d\theta'\int_{-\epsilon_c}^{\epsilon_c}d\epsilon',\qquad(16)$$

where θ' and ϕ' are the polar and azimuthal angles of $\hat{\mathbf{p}}'$. 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 Im $\Sigma(\omega)=-$sign ω the ϵ' integration gives for $\delta_n(H)$

$$\Sigma(\omega) = g^2 N(0) \sum_{\alpha\beta,n} \langle q_\alpha^2 q_\beta^2 \rangle |\langle \Gamma_s | O_{\alpha\beta} | \Gamma_t^n \rangle|^2 \times \int \frac{d\omega'}{2\pi} \frac{2\delta_n}{(\omega-\omega')^2 - \delta_n^2 + i\eta} \times \text{sign}(\omega') 2 \arctan \frac{\epsilon_c}{|\text{Im}\,\Sigma(\omega')|}. \quad (17)$$

We solve the self-consistency equation for the imaginary part Im $\Sigma(\omega)$,

$$\text{Im}\,\Sigma(\omega) = -g^2 N(0) \sum_{\alpha\beta,n} \langle q_\alpha^2 q_\beta^2 \rangle |\langle \Gamma_s | O_{\alpha\beta} | \Gamma_t^n \rangle|^2 \sum_{\rho=\pm 1} \text{sign}[\omega + \rho\delta_n(H)] \arctan \frac{\epsilon_c}{|\text{Im}\,\Sigma[\omega + \rho\delta_n(H)]|}, \quad (18)$$

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

$$|\text{Im}\,\Sigma(\omega)| \le \pi g^2 N(0) \sum_{\alpha\beta,n} \langle q_\alpha^2 q_\beta^2 \rangle |\langle \Gamma_s | O_{\alpha\beta} | \Gamma_t^n \rangle|^2 = \frac{\hbar}{\tau}. \quad (19)$$

It is important to note that 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 $\delta_+(H)$,

$$|\text{Im}\,\Sigma[\delta_+(H)+\eta] - \text{Im}\,\Sigma[\delta_+(H)+\eta]| \ge \frac{\hbar}{\tau}\frac{2}{\pi} \arctan \frac{\epsilon_c \tau}{\hbar}. \quad (20)$$

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

In the limit $\epsilon_c \to \infty$ where $2 \arctan \frac{\epsilon_c}{|\text{Im}\,\Sigma(\omega')|} \to \pi$ the result agrees with that of the non-self-consistent second-order perturbation theory. In this case differentiating the self-energy with respect to ω under the integral and using integration by parts, one finally gets from Eq. (12),

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

$$\chi_Q(H) = \sum_{\alpha\beta,n} \frac{2|\langle \Gamma_s(H) | O_{\alpha\beta} | \Gamma_t^n(H) \rangle|^2}{\delta_n(H)}. \quad (21)$$

The directional average (with respect to polar and azimuthal angles θ and ϕ of $\hat{\mathbf{q}}$) for quadrupolar form factors $\bar{f} = \langle \hat{q}_\alpha^2 \hat{q}_\beta^2 \rangle = \frac{1}{15}$ 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.

TABLE I. Singlet-triplet CEF states, levels, and excitation energies in a magnetic field $\mathbf{H}\|[001]$. Here $\delta_n(H) = E_t^n(H) - E_g(H)$. The eigenstates are given in terms of zero-field singlet-triplet states $|0,0\rangle$ and $|1,\pm\rangle, |1,0\rangle$, respectively ($h = g\mu_B H$).

Eigenstate	$\|\Gamma(H)\rangle$	$E(H)$	$\delta_n(H)$
$\|\Gamma_s(H)\rangle$	$u\|0,0\rangle + v\|1,0\rangle$	$\frac{1}{2}(\Delta - \tilde{\Delta})$	0
$\|\Gamma_t^+(H)\rangle$	$\|1,+\rangle$	$\Delta - h$	$\frac{1}{2}(\Delta + \tilde{\Delta}) - h$
$\|\Gamma_t^0(H)\rangle$	$u\|1,0\rangle - v\|0,0\rangle$	$\frac{1}{2}(\Delta + \tilde{\Delta})$	$\tilde{\Delta}$
$\|\Gamma_t^-(H)\rangle$	$\|1,-\rangle$	$\Delta + h$	$\frac{1}{2}(\Delta + \tilde{\Delta}) + h$

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.*[9,10] 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 $|\Gamma_t^n\rangle$ ($n = +, 0, -$), respectively. The CEF and Zeeman Hamiltonians can be easily mapped to a pseudospin basis[9,10] and then diagonalized. 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 $\mathbf{H}\|[001]$ 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 eperiments.[20]

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

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

$$\tilde{\Delta} = [\Delta^2 + 4\delta^2 h^2]^{1/2}, \quad h \equiv g\mu_B \alpha H. \quad (22)$$

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 $|\Gamma_t^0\rangle$ 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 $|\Gamma_t^\pm\rangle$ have a linear Zeeman splitting of $2h$ that is independent of d. For $\delta \ge 0$ 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 $\delta_+(h_c) = 0$.

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 $\alpha' = \frac{\sqrt{3}}{4}(13-20d^2)$ and $\beta' = \sqrt{35(1-d^2)}$. With their help and defining $\delta' = \alpha'/\beta'$ one obtains the following nonzero matrix elements:

$$|\langle\Gamma_s|O_{yz}|\Gamma_t^+\rangle|^2 = |\langle\Gamma_s|O_{zx}||\Gamma_t^-\rangle|^2 = \frac{1}{2}\beta'^2(u-\delta' v)^2 = |m_Q^-|^2,$$

$$|\langle\Gamma_s|O_{yz}|\Gamma_t^-\rangle|^2 = |\langle\Gamma_s|O_{zx}||\Gamma_t^+\rangle|^2 = \frac{1}{2}\beta'^2(u+\delta' v)^2 = |m_Q^+|^2,$$

$$|\langle\Gamma_s|O_{xy}|\Gamma_t^0\rangle|^2 = \beta'^2 = |m_Q^0|^2. \quad (23)$$

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

$$\chi_Q(H) = \frac{2\beta'^2}{\Delta}\left\{\frac{2\Delta\Delta'}{\Delta'^2-h^2}[1-v^2(1-\delta'^2)] + \frac{\Delta}{\tilde{\Delta}}\right\}, \quad (24)$$

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

$$\chi_Q(H) = \frac{2\beta'^2}{\Delta}\left[\frac{2\Delta^2}{\Delta^2-h^2}[1-v^2(1-\delta'^2)] + 1\right]. \quad (25)$$

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

$$\chi_Q(H) \simeq \frac{2\beta'^2}{\Delta}\left\{3 + [2\delta^2(\delta'^2-3)]\left(\frac{h}{\Delta}\right)^2\right\}. \quad (26)$$

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

$$\frac{m^*}{m} = 1 + g^2 N(0) 3\bar{f}\frac{2\beta'^2}{\Delta}. \quad (27)$$

The states and energies in Table I are nominally derived for $\mathbf{H}\|[001]$. However it was shown in Refs. 9 and 10 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 Δ. This condition is required anyway in the dispersionless case where the calculation is only valid for moderate fields when $\delta_+(h)$ is still large enough.

V. INFLUENCE OF QUADRUPOLE EXCITON DISPERSION ON THE MASS ENHANCEMENT

In Sec. IV we investigated a model of noninteracting local singlet-triplet quadrupole excitations. For appreciably large concentration of Pr ions in the system $Pr_{1-x}La_xOs_4Sb_{12}$, this is no longer justified. Due to effective interactions between the $4f$ states on different sites, the singlet-triplet excitations at Δ acquire a dispersion. Formally this is already included in the self-energy of Eq. (13), provided that the boson propagator for a dispersive mode is used by replacing $\delta_n \to \omega_n(\mathbf{q})$ 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[12]

$$H_Q = \sum_{\langle ij\rangle} K_Q(ij)\mathbf{O}(i)\cdot\mathbf{O}(j), \quad (28)$$

where $\mathbf{O}=(O_{yz},O_{zx},O_{xy})$ is the Γ_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 (AFQ) order from which the Γ_5 symmetry has been infered.[9] Formally H_Q may be obtained from an RKKY-type mechanism in the order of $\sim g^2$ in the coupling constant of H_{AC}. In practice the nearest-neighbor (NN) term K_Q is determined from the experimentally observed dispersion of the quadrupolar excitons $\omega_n(\mathbf{q})$ 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 generalized Holstein-Primakoff approximation[12] the three quadrupolar exciton modes at moderate fields are described by

$$\omega_\pm(\mathbf{q}) = \omega(\mathbf{q}) \mp h, \quad \omega_0(\mathbf{q}) = \omega(\mathbf{q}), \quad (29)$$

where the zero-field dispersion is given by

$$\omega(\mathbf{q}) = \sqrt{A_\mathbf{q}^2 - B_\mathbf{q}^2},$$

$$A_\mathbf{q} = \Delta + zK_Q\gamma_\mathbf{q}, \quad B_\mathbf{q} = -zK_Q\gamma_\mathbf{q}, \quad (30)$$

$$\gamma_\mathbf{q} = \cos\frac{1}{2}q_x \cos\frac{1}{2}q_y \cos\frac{1}{2}q_z. \quad (31)$$

Here $z=8$ is the coordination number and $\gamma_\mathbf{q}$ is the structure function of the bcc cubic lattice of Pr ions with momentum $\mathbf{q}$ measured in r.l.u. ($2\pi/a$). The width of the exciton dispersion is controlled 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 $g_Q \simeq 0.3$ in $PrOs_4Sb_{12}$. The minimum of the dispersion occurs at the bcc zone-boundary wave vector $\mathbf{Q}=(1,0,0)$ (r.l.u.). The zero-field energy is given by $\omega(\mathbf{Q})=\Delta[1-2g_Q]^{1/2}$. 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 $PrOs_4Sb_{12}$. Therefore application of a magnetic field is necessary to achieve a soft mode $\omega_+(\mathbf{Q})=0$ at a critical field h_c. The dispersions in Eq. (31) are approximations where the field dependence of the $\Gamma_s-\Gamma_t^0$ 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 $h_c/\Delta=(1-2g_Q)^{1/2}$ 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 of 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 Sec. III. The main modifications arise from the

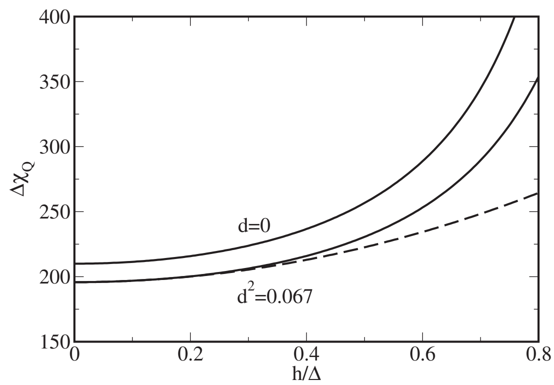

FIG. 2. Static (normalized) quadrupolar susceptibility $\Delta\chi_Q$ as a function of magnetic field with ($d^2=0.067$ or $|d|=0.26$) and without ($d^2=0$) tetrahedral CEF. Full lines correspond to evaluation with Eq. (24) while the dashed line corresponds to the low-field expansion Eq. (26). Approaching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

fact that a more sophisticated approximation for the electron-quadrupolar exciton spectral function is employed. For large cutoff energies $\epsilon_c\to\infty$ one obtains

$$\frac{m^*}{m}=1+g^2N(0)\sum_{\alpha\beta,n}2|\langle\Gamma_s|O_{\alpha\beta}|\Gamma_t^n\rangle|^2\frac{1}{2\pi}\int d\Omega_{\mathbf{q}}\frac{\hat{q}_\alpha^2\hat{q}_\beta^2}{\omega_n(\mathbf{q})}, \tag{32}$$

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

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

$$\chi_Q(H)=\frac{2\beta'^2}{\Delta}\frac{1}{\bar{f}}\Bigg\{[1-v^2(1-\delta'^2)]\times\frac{1}{2\pi}\int d\Omega_{\mathbf{q}}\frac{\Delta\omega(\mathbf{q})}{\omega(\mathbf{q})^2-h^2}\hat{q}_z^2(\hat{q}_x^2+\hat{q}_y^2)+\frac{1}{2\pi}\int d\Omega_{\mathbf{q}}\frac{\Delta}{\omega(\mathbf{q})}\hat{q}_x^2\hat{q}_y^2\Bigg\}. \tag{33}$$

Here the first and second terms are due to the virtual excitations of $\omega_\pm(\mathbf{q})$ and $\omega_0(\mathbf{q})$ 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 and E_t^0 level repulsion implied in the dispersions of Eq. (29). The above expression for the mass enhancement has to be evaluated numerically due to the BZ integrations. This will be discussed in Sec. VI. But we may nevertheless gain some qualitative insights by simple approximations to these integrals in the zero-field case. For that purpose we expand $\omega(\mathbf{q})$ around one of the six equivalent zone-boundary X points with $\mathbf{Q}=(\pm1,0,0)$, etc. Then one obtains an isotropic approximate dispersion given by

$$\omega(\mathbf{q}')^2=\omega_{\mathbf{Q}}^2+\omega_0^2(\pi q')^2, \tag{34}$$

where $\mathbf{q}'=\mathbf{Q}-\mathbf{q}$ is the momentum vector counted from the X point and q' is its length. Furthermore $\omega_{\mathbf{Q}}=\Delta[1-2g_Q]^{1/2}=h_c$ and $\omega_0=\sqrt{g_Q}\Delta$. On approaching the critical $g_Q^c=\frac{1}{2}$ the soft-mode frequency $\omega_{\mathbf{Q}}$ vanishes. In this limit the integral in Eq. (32) may easily be evaluated. The approximate Fermi-surface geometry of $PrOs_4Sb_{12}$ shows that $p_F\simeq\frac{1}{\sqrt{2}}$ (r.l.u.) or $2p_F=\sqrt{2}$ (Fig. 1). Therefore $2p_F>Q$ which means that the minimum in $\omega(\mathbf{q})$ is included in the domain of the momentum integrals in Eqs. (32) and (33). We restrict the latter to the sphere around the minimum at $\mathbf{Q}$ (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),

$$\frac{m^*}{m}\simeq 1+g^2N(0)\left(\frac{q_c'^4}{4\pi}\right)\frac{2\beta'^2}{\omega_0}, \tag{35}$$

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 $\omega_{\mathbf{Q}}\to 0$ at the zone boundary, there is no divergence in the mass renormalization. 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 $\hat{q}_\alpha^2\hat{q}_\beta^2$ 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 renormalization diverges when $\Delta\to 0$ because this corresponds to a softening in the whole BZ. Thus we conclude from Eqs. (27) and (35) that the inclusion of a mode dispersion removes the problem of singular mass renormalization, m^*/m stays finite for all coupling constants g_Q, even when $g_Q=g_Q^c$ when the exciton frequency becomes soft at $\mathbf{Q}$. 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. (33).

VI. NUMERICAL RESULTS AND DISCUSSION

We first discuss the mass enhancement in the large bandwidth approximation $\epsilon_c\to\infty$ for dispersionless undamped CEF excitations. The absolute value of $m^*(h)/m=1$

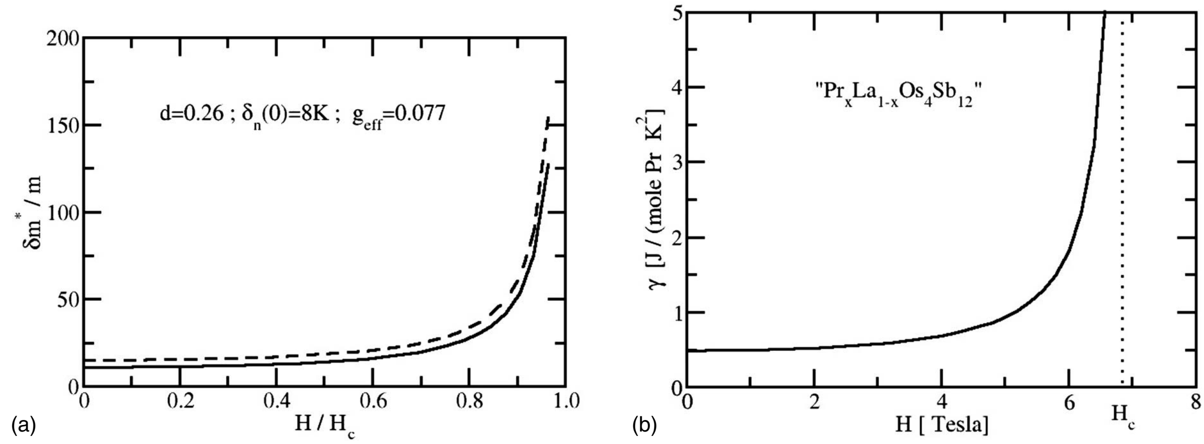

FIG. 3. Left (a): variation with magnetic field of $\delta m^*/m$ calculated from the self-consistent calculation for finite bandwidth $\epsilon_c = 20\delta_n(0)$ (full line) and $\epsilon_c \to \infty$ (broken line). Right (b): variation with magnetic field of the specific-heat coefficient γ calculated self-consistently for $\epsilon_c = 20\delta_n(0)$.

$+\delta m^*/m$ is determined by the effective coupling constant $\tilde{g} = g^2 N(0)$. Approximating $N(0) \sim 1/2W$ ($2W$=bandwidth) this may be written as $\tilde{g}/\Delta = \lambda^2(W/\Delta)$. Here we introduced the dimensionless quadrupolar coupling constant $\lambda = gN(0)$. Assuming typical values of $W=1$ eV, $\lambda \simeq 0.02$, and using $\Delta = 8$ K we obtain $\tilde{g}/\Delta\bar{f} \simeq 0.077$ as the size of the effective coupling for the quadrupolar mass enhancement mechanism. Using $d^2=0.067$ and hence $\beta'^2 \simeq 32.6$ we obtain a zero-field enhancement of $m^*/m \simeq 16$. 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 realized in $Pr_{1-x}La_xOs_4Sb_{12}$, the level repulsion of Γ_s and Γ_t^0 is also weak, and therefore the Γ_t^+ level crosses the Γ_s ground state at a critical field $h_c = \Delta/(1-\delta^2)$ in the dispersionless case. The decrease in the excitation gap for $h < h_c$ and the field dependence of matrix elements lead to a field dependence of $\chi_Q(h)$ [Eq. (24)] 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 $PrOs_4Sb_{12}$. This case resembles more that of pure metallic 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 bandwidth. Self-consistency leads to an overall reduction in the mass renormalization as can be seen from Fig. 3.

The (unphysical) divergence of the mass enhancement close to the critical magnetic field is an artifact 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 Δ. An applied field of critical strength therefore leads to a softening of $\omega(\mathbf{q})$ only in the restricted phase space around the AFQ ordering vector $\mathbf{Q}$. Consequently the mass renormalization will be finite even at the critical field h_c for AFQ order when $\omega(\mathbf{Q})=0$. This is shown in Fig. 4(a) for various effective quadrupolar coupling strengths g_Q. For $g_Q = 0$ the mass renormalization at h_c would diverge as in Fig. 2 because the excitation energy becomes soft for all $\mathbf{q}$ vectors. For small g_Q and dispersive width the softening appears only around $\mathbf{Q}$ [Fig. 4(b)] 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 $\mathbf{Q}$ [see Fig. 4(b)]. For the value $g_Q=0.3$ appropriate for $PrOs_4Sb_{12}$ the dispersion is sufficiently large to suppress the field dependence of $\delta m^*/m$ as depicted in Fig. 4(a) (full line).

An alternative presentation of results for the dispersive case is shown in Fig. 5. The mass enhancement is shown as a function of quadrupolar coupling g_Q (approximately the dispersive width) for the two limiting cases $h=0$ and $h = h_c(g_Q)$. For large g_Q and dispersion the field variation in $\delta m^*/m$ 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 $g_Q \to 0$. The arrow corresponds to the proper value of g_Q for $PrOs_4Sb_{12}$, 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

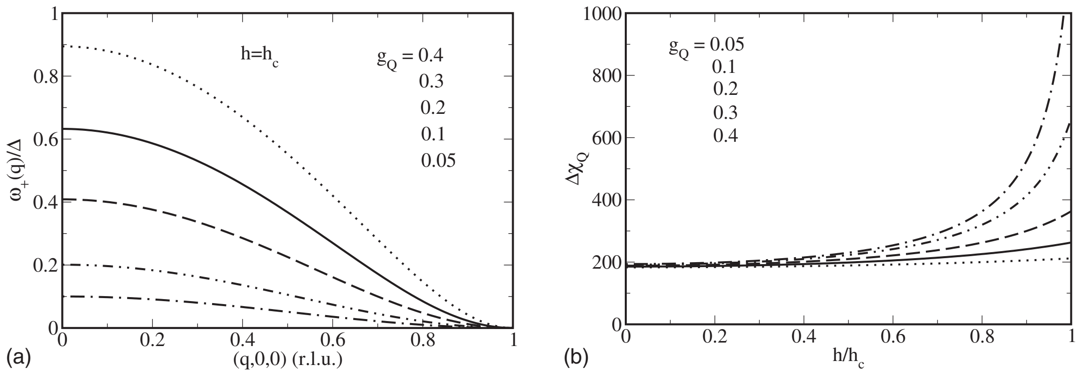

FIG. 4. Left (a): dispersion of lowest triplet quadrupolar exciton $\omega_+(\mathbf{q})$ at the critical field $h_c(g_Q)$ where $\omega_+(\mathbf{Q})$ becomes soft ($\mathbf{Q}$ in units of $2\pi/a$). As g_Q is reduced the dispersion becomes flat increasing the phase space for low-energy conduction-electron 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 renormalization close to h_c is large due to flat $\omega_+(\mathbf{q})$ dispersion. For larger g_Q the dispersion becomes stronger and $\omega_+(\mathbf{q})$ softens only in the vicinity of $\mathbf{Q}=(0,0,1)$, 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 $PrOs_4Sb_{12}$ little field dependence of $\delta m^*/m$ remains.

excitations. This model has some relevance for the heavy fermion superconductor $PrOs_4Sb_{12}$ 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 quasiparticle mass renormalization which may well be the source of the large thermal and dHvA effective masses observed in $PrOs_4Sb_{12}$. A hybridization mechanism between conduction and $4f$ electrons can be ruled out since the Fermi surface of $PrOs_4Sb_{12}$ is identical to that of $LaOs_4Sb_{12}$ which advocates for fully localized $4f$ electrons in Pr. Indeed the well-defined CEF excitations seen in INS (Ref. 21) support this view.

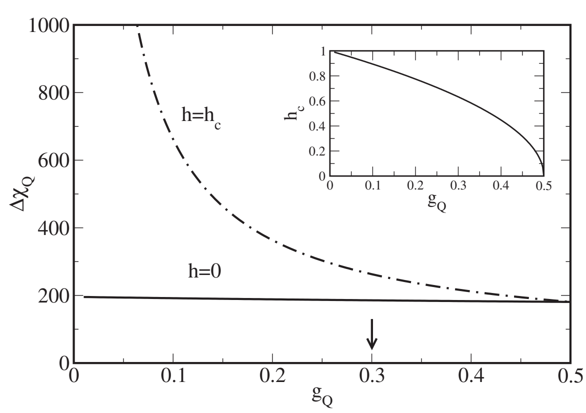

FIG. 5. Quadrupolar susceptibility $\Delta\chi_Q \sim \delta m^*/m$ as a function of effective intersite coupling g_Q. For zero field the intersite coupling or dispersive width of $\omega(\mathbf{q})$ has little effect on the mass enhancement. It actually decreases slightly when g_Q^c=0.5 is approached despite the appearance of the soft mode $\omega(\mathbf{Q})\to 0$. At the critical field h_c the dispersion has much stronger influence; when the latter becomes small (decreasing g_Q) the mass enhancement at h_c strongly increases. For $PrOs_4Sb_{12}$ (g_Q=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 $d^2 \simeq 0$.

If aspherical Coulomb scattering of conduction electrons plays a role in the mass enhancement, one should expect a field dependence of the latter because the triplet excited state splits in the field. For small enough tetrahedral CEF characterized 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 states increases with field, leading to a decrease in the mass enhancement, similar to that observed in Pr metal where the mass renormalization 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 artifact of the dispersionless model, both in the perturbative and self-consistent 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 $\mathbf{Q}$ of incipient field-induced AFQ order. For a quadrupolar coupling constant g_Q=0.3 corresponding to $PrOs_4Sb_{12}$, 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 $PrOs_4Sb_{12}$ 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 $Pr_{1-x}La_xOs_4Sb_{12}$. 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 be progressively decrease. This means that the field dependence of effective masses will become more pronounced according to Figs. 4

and 5. 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 Ce sites. There should however be an intermediate concentration region for x where the field dependence is pronounced (g_Q small) and the $\gamma(x)$ is 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 $Pr_{1-x}La_xOs_4Sb_{12}$ is systematically investigated and analyzed. It may hold important clues to the microscopic origin of the large effective mass in the concentrated compound $PrOs_4Sb_{12}$.

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