Strong-coupling theory of magnetic-exciton-mediated superconductivity in UPd₂Al₃

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There is compelling evidence from inelastic-neutron-scattering and tunneling experiments that the heavyfermion superconductor UPd₂Al₃ can be understood as a dual system consisting of magnetic excitons, arising from crystal-field-split U⁴⁺ levels, coupled to delocalized *f* electrons. We have computed the superconducting transition temperature and the mass renormalization arising from a dual model with maximal spin anisotropy using a strong-coupling approach. We find an instability to two possible opposite-spin-pairing states with evenor odd-parity gap functions. Each has a line node perpendicular to the *z* direction, in agreement with NMR relaxation-rate, specific-heat and thermal-conductivity measurements. In addition, both have total spin component *S_z*=0, compatible with the observation of a pronounced Knight shift and *H_{c2}* Pauli limiting. For parameter values appropriate to UPd₂Al₃, the calculated superconducting transition temperature and mass renormalization agree well with experiment for representative values of the coupling constant.

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I. INTRODUCTION

Heavy-fermion superconductors are complicated materials which possess a fascinating and rich variety of physical properties and which have stimulated the creation of an equally diverse range of theories in an effort to understand them. These materials are dominated by strong electronic correlations giving rise to magnetic and superconducting instabilities. Accordingly, most theories ignore phononic degrees of freedom.

One of the most extensively studied models of heavyfermion superconductivity is a phenomenological model based on the exchange of spin fluctuations between heavy quasiparticles. The latter originate in periodic resonant scattering of conduction electrons close to the Fermi level as described within the Kondo lattice model of Ce compounds.^{1,2} Pairing via spin fluctuations can be considered as a one-component model to the extent that the spin fluctuations originate in the system of heavy quasiparticles and any interaction between conduction electrons and more localized electrons, or conduction electrons and phonons, is neglected. In spite of its simplicity this model has proved to be very useful in our attempts to understand heavy-fermion compounds on the border of magnetic long-range order, such as CePd₂Si₂,³ CeIn₃,³ and CeRhIn₅.⁴ Its success probably derives from the fact that the spin-fluctuation-induced effective interaction tends to dominate all other channels of interaction when a material is tuned close to the border of magnetism by doping or, as in the Ce compounds mentioned above, by applying pressure. The spin-fluctuation mechanism in the Kondo lattice may also be appropriate for the Cebased heavy-fermion superconductors at ambient pressure, CeCu₂Si₂ and CeCoIn₅.^{4,5}

It has become increasingly evident that the Kondo-lattice model is, however, not adequate in the case of heavyquasiparticle formation in some uranium heavy-fermion compounds,^{6,7} where 5f electrons are partly localized and partly itinerant. The former occupy crystalline-electric-field (CEF) split $5f^2$ states. The latter, more itinerant, 5f electrons have a strongly enhanced effective mass due to a coupling to virtual excitations between CEF states. This notion of heavyquasiparticle formation is especially appropriate for UPd₂Al₃ and it is the starting point of our theoretical model for heavyfermion superconductivity in this compound. Specifically, we consider an effective pairing mechanism which is based on the virtual exchange of collective CEF excitations known as "magnetic excitons." They are propagating bosonic modes in contrast to the overdamped modes which give rise to pairing in the itinerant spin-fluctuation model.

The interplay of conduction electrons and CEF excitations is a well studied subject. Initially, research was focused in two areas:⁸ transport anomalies and superconducting pair breaking or enhancement due to conduction-electron scattering from dilute CEF-split impurities; and periodic lattices of CEF ions interacting with one another via conductionelectron polarization. In the latter case attention was focused on collective effects within the CEF system due to the RKKY interactions mediated by itinerant electrons. Later, White and Fulde⁹ showed that the inverse effect, viz. conduction-electron mass enhancement via virtual emission and absorption of magnetic excitons, is also important. They demonstrated that this mechanism explains the enhanced effective electron masses in praseodymium metal.

It is then natural to ask whether the exchange of magnetic excitons between quasiparticles can also mediate superconductivity. Until recently, no theoretical work had been carried out to answer this question, mainly for two reasons. First, when the effects on superconductivity of paramagnetic impurities with CEF-split energy levels were studied in detail for the s-wave case, ¹⁰ it was found that for rare-earth ions the pair-breaking transition matrix elements usually dominate the pair-enhancing matrix elements. Second, no good example of a superconducting compound with this dual nature, i.e., magnetic excitons arising from localized f electrons

coupled to delocalized f electrons, was known. Recently, however, evidence has been accumulating that the heavy-fermion compound UPd₂Al₃ (T_N =14.3 K, T_c =1.8 K, see Ref. 11) is the first example where such a mechanism is responsible for superconductivity. Since this mechanism is pair breaking in the s-wave channel (as was already known from the impurity models mentioned above), the superconducting gap function must change sign as a function of **p** (i.e., the gap must have a node) as is the case with spin-fluctuation-mediated superconductivity.

An early indication that UPd_2Al_3 is a localizeddelocalized f system came from magnetic-susceptibility measurements. Grauel *et al.*¹² measured the dc magnetic susceptibility in UPd_2Al_3 and argued that the anisotropy which they observed arose from a tetravalent configuration of the uranium ions, i.e., U^{4+} (5 f^2). Since then Knight-shift¹³ and optical¹⁴ measurements have supported the notion that UPd_2Al_3 contains both localized and delocalized f electrons.

However, the most direct evidence in favor of the dualsystem hypothesis comes from inelastic neutron scattering (INS) and tunneling experiments. A dispersive crystal-field excitation (magnetic exciton) was observed by Mason and Aeppli¹⁵ in INS experiments within the antiferromagnetic (AF) phase. Later higher-resolution experiments¹⁶⁻¹⁸ revealed a resonance feature which appears in the INS spectrum upon entering the superconducting state. This result demonstrates that a strong interaction exists between the localized and delocalized components of the *f*-electron system. experiments¹⁹ Pioneering tunneling performed on UPd₂Al₃-AlO_y-Pb tunneling junctions allowed experimenters to view, for the first time, the tunneling density of states (DOS) of a heavy-fermion superconductor. Strong-coupling features appear in the DOS around 1 meV close to the superconducting gap energy reinforcing the view that the exchanged bosons are the magnetic excitons. Taken together, INS and tunneling experiments led Sato and co-workers²⁰ to the conclusion that superconductivity arises in this material from an effective interaction between itinerant electrons mediated by magnetic excitons. Using a model two-component Hamiltonian they were able to explain qualitative features of the INS scattering spectrum and the superconducting tunneling spectrum.

Subsequently, the origin of the magnetic excitons and their global dispersion, as measured in Ref. 15, was investigated in more detail.²¹ Using an *xy*-type interaction, where only the σ_{\pm} components of the conduction-electron spin couples to the magnetic excitons, the effective nonretarded pair potential was derived. The gap equations were solved within a weak-coupling approach demonstrating that the highest T_c is obtained by an odd-parity state. In this model, however, the structure of the pairing amplitudes in spin space is complicated, making it unsuitable for going beyond the nonretarded approximation.

In this paper, therefore, we investigate an alternative model with a simplified interaction between localized and itinerant 5f electrons. This interaction is of the Ising type, i.e., only the σ_z component of the conduction-electron spin can scatter magnetic excitons. In this case, the gap equations naturally divide into those for equal- and opposite-spin pairing, in contrast to the usual "singlet" and "triplet" classifica-

tion that arises in spin-rotation-symmetric models. In the present work we treat this simplified dual model for UPd₂Al₃ in a more sophisticated strong-coupling approach using a mapping to an electron-boson Hamiltonian and solving the Éliashberg equations for the frequency- and momentum-dependent self-energy and gap functions. The level scheme which we use here, and which gives rise to the Ising-type interaction, is quite realistic: recent band-structure calculations based on this level scheme account well for experimental de Haas-van Alphen frequencies in UPd₂Al₃.⁷ We demonstrate that our model can yield a superconducting transition temperature and a mass renormalization which are consistent with experiment for reasonable values of the coupling constant.

II. MODEL

Band structure calculations based on the supposition that two of the three uranium 5*f* electrons are localized reproduce the observed de Haas-van Alphen frequencies in UPd₂Al₃ very well.⁷ These calculations suggest a level scheme for the localized U 5*f* states which we adopt here. According to the *jj*-coupling scheme the U⁴⁺(5*f*²) ions have total angular momentum *J*=4. The twofold degeneracy of the ionic ground state is lifted by a crystalline electric field. We consider only the excitation between the non-degenerate ground state $|\Gamma_4\rangle$ $=\frac{1}{\sqrt{2}}(|J_z=3\rangle - |J_z=-3\rangle)$ and the first excited state $|\Gamma_3\rangle = \frac{1}{\sqrt{2}}(|J_z=3\rangle + |J_z=-3\rangle)$. The CEF energy splitting is of the order Δ ≈ 6 meV, as obtained from fitting the magnetic-exciton dispersion obtained from INS measurements¹⁵ to theoretical results.²¹

The dual model of 5f electrons may also exhibit a magnetic instability which is completely dominated by the localized electrons $(T_N \ll E_F)$. Because of the singlet-singlet splitting the ordered moments must be of the induced moment variety: non-zero magnetic moments arise because the states $|\Gamma_3\rangle$ and $|\Gamma_4\rangle$ are mixed by effective interionic exchange interactions. Indeed this mechanism was proposed in Ref. 21 as the origin of the AF order observed at $T_N = 14.3$ K. With the work in Ref. 21 in mind, we take the view here that the underlying AF order is not an important consideration in the description of heavy-fermion superconductivity in UPd₂Al₃. AF order mainly leads to a folding down of the conduction bands into the AF Brillouin zone. Also, the magnetic-exciton dispersion in the ordered phase is not appreciably different from that in the paramagnetic phase, due to the fact that $T_N \ll \Delta$ ²¹ Furthermore, in mean-field theory, the AF order parameter will only modify the superconducting T_c to the extent that the conduction-electron states are reconstructed close to the AF Bragg planes.

Accordingly we consider a three-dimensional lattice of localized $5f^2$ CEF states and itinerant f electrons. In the subspace $\{|\Gamma_3\rangle, |\Gamma_4\rangle\}$ we may write the CEF part of the Hamiltonian as

$$H_{\rm CEF} = \Delta \sum_{i} S_{iz},\tag{1}$$

where **S** denotes a pseudospin $(S=\frac{1}{2})$. In this representation we interpret the CEF ground state as having $S_z=-\frac{1}{2}$ and en-

ergy $-\Delta/2$ and the excited CEF state as having $S_z = \frac{1}{2}$ and energy $\Delta/2$. In the pseudospin representation, the only nonzero component of the physical total angular momentum **J** is the J_z component:

$$J_z = \gamma \begin{pmatrix} 0 & \frac{1}{2} \\ \frac{1}{2} & 0 \end{pmatrix} = \gamma S_x, \qquad (2)$$

where $\gamma = 6$. Then the full two-component Hamiltonian may be written as

$$H = \sum_{\mathbf{p}\sigma} \epsilon_{\mathbf{p}\sigma} c_{\mathbf{p}\sigma}^{\dagger} c_{\mathbf{p}\sigma} + \Delta \sum_{i} S_{iz} - J \sum_{i\delta} S_{x}^{i} S_{x}^{i+\delta} - I \sum_{i} \sigma_{iz} S_{ix}.$$
 (3)

The third term in H is a nearest-neighbor superexchange interaction between localized 5f states. The last term is the exchange interaction between localized 5f CEF states and the delocalized 5f conduction electrons; the conductionelectron spin operator is

$$\boldsymbol{\sigma}_{i} = \boldsymbol{\psi}_{\alpha}^{\dagger}(\mathbf{r}_{i}) \boldsymbol{\sigma}_{\alpha\beta} \boldsymbol{\psi}_{\beta}(\mathbf{r}_{i}). \tag{4}$$

After carrying out a Holstein-Primakoff-like transformation (valid at low temperatures $T \ll \Delta$) and an additional Bogoliubov transformation involving the resulting bosons (see Appendix A), *H* takes the form

$$H = \sum_{\mathbf{p}\sigma} \epsilon_{\mathbf{p}} c^{\dagger}_{\mathbf{p}\sigma} c_{\mathbf{p}\sigma} + \sum_{\mathbf{q}} \omega_{\mathbf{q}} (\alpha^{\dagger}_{\mathbf{q}} \alpha_{\mathbf{q}} + 1/2) - I \int d\mathbf{r} \psi^{\dagger}_{\alpha}(\mathbf{r}) \sigma^{z}_{\alpha\beta} \psi_{\beta}(\mathbf{r}) \phi(\mathbf{r})$$
(5)

where

$$\phi(\mathbf{r}) = \frac{1}{\sqrt{V}} \sum_{\mathbf{q}} \frac{1}{2} \lambda_{\mathbf{q}} (\alpha_{\mathbf{q}} + \alpha_{-\mathbf{q}}^{\dagger}) e^{i\mathbf{q}\mathbf{r}}, \quad \lambda_{\mathbf{q}}^2 = \frac{\Delta}{\omega_{\mathbf{q}}}.$$
 (6)

 $\omega_{\mathbf{q}}$ is the dispersion of those bosons which have creation and destruction operators $\alpha_{\mathbf{q}}^{\dagger}$ and $\alpha_{\mathbf{q}}$, respectively (see Appendix A).

We define the electron and magnetic-exciton Green's functions as follows:

$$G_{\alpha\beta}(\mathbf{r}-\mathbf{r}',\tau-\tau') = -\langle T\psi_{\alpha}(\mathbf{r},\tau)\overline{\psi}_{\beta}(\mathbf{r}',\tau')\rangle, \qquad (7)$$

$$D(\mathbf{r} - \mathbf{r}', \tau - \tau') = -\langle T\phi(\mathbf{r}, \tau)\phi(\mathbf{r}', \tau')\rangle.$$
(8)

Note that $\phi(\mathbf{r}) \sim S_x(\mathbf{r})$, as can be seen immediately by comparing the terms $\sim I$ in Eqs. (3) and (5), and so *D* is essentially a pseudospin susceptibility. Furthermore, as in the theory of phonon-mediated superconductivity, $\phi(\mathbf{r})$ is real, $\phi(\mathbf{r}) = \phi^{\dagger}(\mathbf{r})$, and commutes with itself, $[\phi(\mathbf{r}), \phi(\mathbf{r}')] = 0$, conditions which allow us to use Wick's theorem. We assume that we may write the superconducting order parameter $\Phi_{\alpha\beta}(\mathbf{p}, i\omega_n)$ as the product of a part which contains the momentum and frequency dependence $\Phi(\mathbf{p}, i\omega_n)$ and a part that represents the spin state of the paired quasiparticles $|\chi\rangle$,

$$\Phi_{\alpha\beta}(\mathbf{p},i\omega_n) = \Phi(\mathbf{p},i\omega_n) \langle \alpha\beta | \chi \rangle. \tag{9}$$

Then the Éliashberg equations for the conduction-electron self-energy $\Sigma(\mathbf{p}, i\omega_n)$ and gap function $\Phi(\mathbf{p}, i\omega_n)$ which follow from the Hamiltonian Eq. (5) are

$$\Sigma(\mathbf{p}, i\omega_n) = \frac{T}{N} \sum_{\mathbf{p}'\omega_n'} K(\mathbf{p} - \mathbf{p}', i\omega_n - i\omega_n') G(\mathbf{p}', i\omega_n'), \quad (10)$$

$$\Phi(\mathbf{p}, i\omega_n) = p \frac{T}{N} \sum_{\mathbf{p}'\omega_n'} K(\mathbf{p} - \mathbf{p}', i\omega_n - i\omega_n')$$
$$\times |G(\mathbf{p}', i\omega_n')|^2 \Phi(\mathbf{p}', i\omega_n').$$
(11)

N is the total number of lattice sites. The kernel *K* is given by

$$K(\mathbf{q}, i\nu_n) = -I^2 D^0(\mathbf{q}, i\nu_n) = \left(\frac{I^2 \Delta}{2}\right) \frac{1}{\nu_n^2 + \omega_\mathbf{q}^2}.$$
 (12)

We have decided not to renormalize D^0 by the interaction Ibetween magnetic excitons and electrons. This interaction is, however, already included in D^0 to the extent that we model the exciton dispersion ω_q by the true experimental dispersion. A particularly noteworthy feature of this formulation is that the effective interaction is dominated by its static part and is strongly peaked in \mathbf{q} space at the antiferromagnetic wave vector $\mathbf{Q} = (0, 0, \pi/c)$. This strong dependence on wave vector contrasts with the practically wave-vectorindependent interaction in the usual phonon-mediated superconductivity. The difference arises because, in the phonon problem, the quantity corresponding to λ_q has an additional factor, $\omega_{\mathbf{q}}$, leading to a wave-vector-independent static phonon propagator. A strong interaction between the collective modes of the localized moments and the heavy conduction electrons at $\mathbf{Q} = (0, 0, \pi/c)$ has actually been observed^{17,18} in UPd₂Al₃. The electron Green's function is related to the electron self-energy via the Dyson equation

$$G^{-1}(\mathbf{p}, i\omega_n) = i\omega_n - (\boldsymbol{\epsilon}_{\mathbf{p}} - \boldsymbol{\mu}) - \Sigma(\mathbf{p}, i\omega_n).$$
(13)

The prefactor *p* in Eq. (11) is the expectation value of the Ising spin-spin interaction $\hat{\sigma}^z \hat{\sigma}^z$ in the spin part $|\chi\rangle = |S, S_z\rangle$ of the pair wave function (**S** here should not be confused with the pseudospin introduced earlier):

$$p = \langle \chi | \hat{\sigma}^z \hat{\sigma}^z | \chi \rangle. \tag{14}$$

In the opposite-spin pair (OSP) states,

$$|\chi\rangle = \begin{cases} \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \\ \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle), \end{cases}$$
(15)

p reduces to $p = \langle \uparrow \downarrow | \hat{\sigma}^z \hat{\sigma}^z | \uparrow \downarrow \rangle = -1$. On the other hand, for each of the equal-spin pair (ESP) states, $|\chi\rangle = |\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$, *p* is +1. Note that spin rotational symmetry is broken in a maximal (Ising-type) way in Eq. (11). Consequently the usual classification of pairing states into an (even-parity) singlet with *S*=0 and an (odd-parity) triplet with *S*=1 is not appropriate. In our model, the $|1,0\rangle$ pair state no longer has

TABLE I. Spin and orbital structure of the possible gap functions which are solutions of the Éliashberg equations for the dual model of UPd_2Al_3 used in this paper.

р	$ \chi\rangle$ = $ S,S_z\rangle$	D_{6h} repres.	Spin pairing	$\Phi(p_z)$
-1	$ 0,0\rangle = \frac{1}{\sqrt{2}}(\uparrow\downarrow\rangle - \downarrow\uparrow\rangle)$	$\Gamma_1^+(A_{1g})$	OSP	$\cos(cp_z)$
-1	$ 1,0\rangle = \frac{1}{\sqrt{2}}(\uparrow\downarrow\rangle + \downarrow\uparrow\rangle)$	$\Gamma_1^-(A_{1u})$	OSP	$sin(cp_z)$
+1	$ 1,\pm1\rangle = \uparrow\uparrow\rangle, \downarrow\downarrow\rangle$	$\Gamma_1^-(A_{1u})$	ESP	$\sin(2cp_z)$

the same energy as that of the $|1, \pm 1\rangle$ pair states. The result is a different classification into an ESP doublet and two OSP singlet states belonging to irreducible representations of the D_{6h} group (Table I). We refer the reader to Appendix B where it is shown how the prefactor p arises in Eq. (11).

Finally, the conduction-band filling, defined as the ratio of the number of electrons to the number of lattice sites, is

$$n = \frac{1}{N} \sum_{\mathbf{p}} n_{\mathbf{p}},\tag{16}$$

where $n_{\mathbf{p}}$ is the quasiparticle occupation factor:

(

$$n_{\mathbf{p}} = n_{\mathbf{p}\uparrow} + n_{\mathbf{p}\downarrow} = 2T \sum_{\omega_n} G(\mathbf{p}, i\omega_n).$$
(17)

Experimentally, the magnetic exciton has its strongest dispersion ω_q along the *z* direction. Neglecting the weaker dispersion in the *x*-*y* plane allows us to reduce the threedimensional problem to one dimension:

$$\Sigma(p_z, i\omega_n) = \frac{T}{N_z} \sum_{p'_z \omega'_n} K(p_z - p'_z, i\omega_n - i\omega'_n)$$
$$\times \int \frac{d\mathbf{p}'_\perp}{(2\pi)^2} G(\mathbf{p}'_\perp, p'_z, i\omega'_n), \qquad (18)$$

$$\Phi(p_z, i\omega_n) = p \frac{T}{N_z} \sum_{p'_z \omega'_n} K(p_z - p'_z, i\omega_n - i\omega'_n) \Phi(p'_z, i\omega'_n)$$
$$\times \int \frac{d\mathbf{p}'_\perp}{(2\pi)^2} |G(\mathbf{p}'_\perp, p'_z, i\omega'_n)|^2, \tag{19}$$

$$n = \frac{1}{N_z} \sum_{p_z} A \int \frac{d\mathbf{p}_\perp}{(2\pi)^2} n_{\mathbf{p}},$$
 (20)

provided we can carry out the integrals along the perpendicular direction analytically. Here, N_z is the number of lattice sites in the z direction and A is the area of the (hexagonal) unit cell in the plane. We model the exciton dispersion by

$$\omega(q_z) = \omega_{ex}[1 + \beta \cos(q_z)]; \quad 0 < \beta \simeq 1.$$
 (21)

This form of the dispersion along the *z* direction describes qualitatively the observed excitation branch.¹⁵ Here $2\beta\omega_{ex} \approx 8 \text{ meV}$ is the overall dispersion width and $(1-\beta)\omega_{ex} \approx 1 \text{ meV}$ is the exciton gap at the AF wave vector $\mathbf{Q} = (0,0,\pi/c)$. The quantity ω_{ex} is a characteristic exciton energy. We choose $\beta=0.8$ and $\omega_{ex}\approx 5 \text{ meV}\approx 60 \text{ K}$. The detailed RPA theory of the magnetic-exciton dispersion and a

fit to the INS data of Ref. 15 was given in Ref. 21.

That sheet of the Fermi surface of UPd₂Al₃ thought to be most important in bringing about heavy-fermion behavior²¹ has the shape of a corrugated cylinder with its axis aligned along the z direction of the hexagonal lattice. We therefore choose to model the electron dispersion as the sum of a strongly dispersive part in the plane $\epsilon_{\mathbf{p}_{\perp}}$ and a weakly dispersive part in the z direction ϵ_{p_z} . We approximate the hexagonal unit cell in the plane (p_x, p_y) by a circle with radius p_0 , chosen so that the hexagon and circle have the same area. (The areas must be the same in order that the maximum value of the band filling is two in both cases.) Then, assuming a parabolic dispersion in the plane $\epsilon_{\mathbf{p}_{\perp}} = \epsilon_{\perp} w^2 \ (0 < w$ $=p_{\perp}/p_0 < 1$), we may carry out the integrals in Eqs. (18)–(20) analytically. In the following, when not explicitly stated, we measure all energies, temperatures and frequencies in units of ϵ_{\perp} . The reduced Éliashberg equations are

$$\Sigma(p_z, i\omega_n) = \frac{T}{N_z} \sum_{p'_z \omega'_n} K(p_z - p'_z, i\omega_n - i\omega'_n) G_z(p'_z, i\omega'_n),$$
(22)

$$\lambda(T)\Phi(p_z, i\omega_n) = p \frac{T}{N_z} \sum_{p'_z \omega'_n} K(p_z - p'_z, i\omega_n - i\omega'_n)$$
$$\times M_z(p'_z, i\omega'_n)\Phi(p'_z, i\omega'_n), \qquad (23)$$

$$n = 1 - \frac{2T}{N_z} \sum_{p_z, \omega_n > 0} \ln \left[\frac{(z'-1)^2 + (z'')^2}{(z')^2 + (z'')^2} \right],$$
(24)

where

$$K(q_{z}, i\nu_{n}) = \frac{g}{(\omega_{q_{z}}/\omega_{ex})^{2} + (\nu_{n}/\omega_{ex})^{2}},$$
(25)

$$G'_{z}(p'_{z},i\omega_{n}) = -\frac{1}{2} \ln \left[\frac{(z'-1)^{2} + (z'')^{2}}{(z')^{2} + (z'')^{2}} \right],$$
 (26)

$$G_{z}''(p_{z}',i\omega_{n}) = -\left\{\tan^{-1}\left(\frac{1-z'}{z''}\right) - \tan^{-1}\left(-\frac{z'}{z''}\right)\right\},\quad(27)$$

$$M_{z}(p_{z}',i\omega_{n}) = \frac{1}{z''} \left\{ \tan^{-1} \left(\frac{1-z'}{z''} \right) - \tan^{-1} \left(-\frac{z'}{z''} \right) \right\}.$$
 (28)

and

$$\lambda(T) = 1 \text{ for } T = T_c. \tag{29}$$

We have lumped together numerical prefactors and the original coupling constant I into a new coupling constant g having dimensions of energy:

$$g = \frac{I^2 \Delta}{2} \left(\frac{1}{2c} \frac{p_0^2}{2\pi} \right) \frac{1}{\omega_{ex}^2},\tag{30}$$

where c is the lattice constant in the z direction. The value of g is not calculated; rather it is considered a model parameter motivated by experiment. The complex number z has a real part

$$z' = -\left[\epsilon_{p'_{z}} - \mu + \Sigma'(p'_{z}, i\omega_{n})\right]$$
(31)

and an imaginary part

$$z'' = \omega_n - \Sigma''(p'_z, i\omega_n). \tag{32}$$

We choose a simple tight-binding form for $\epsilon_{p'_z}$

$$\epsilon_{p_z'} = \epsilon_{\perp} \left(\frac{\alpha}{2}\right) \cos(p_z'); \quad \alpha \ll 1,$$
(33)

where α determines the degree of corrugation of the FS cylinder along the z axis.

The momentum convolutions in Eqs. (22) and (23) were evaluated with the aid of a fast-Fourier-transform algorithm on a 32×32 lattice. The corresponding frequency sums were carried out using the renormalization group technique of Pao and Bickers²² which allows a considerable reduction of the computational effort. Between 240 and 480 Matsubara frequencies were kept at each stage of the renormalization group procedure. The renormalization procedure was started at a temperature $T_0=0.01\epsilon_{\perp}$ and the frequency sum cut-off used was $\Omega_c \approx 15\epsilon_{\perp}$. The renormalization procedure restricts us to discrete temperatures so that the point at which the condition in Eq. (29) is met must be determined by interpolation. The discrete temperatures were sufficiently close that a linear interpolation was adequate.

III. RESULTS AND DISCUSSION

To begin with a comment is in order regarding vertex corrections to the self-consistent Éliashberg theory presented in Sec. II. In Migdal's analysis of the electron-phonon problem these corrections were shown to scale as the ratio $\omega_D/E_F \ll 1$, where ω_D is the Debye energy. Although ω_{ex} is of the same order of magnitude as ω_D , one might be inclined to think that E_F in UPd₂Al₃ is comparable to ω_{ex} because of the low velocity of the heavy quasiparticles. However, this is not the case. Vertex corrections involve an integration over momentum transfers of the order of the Fermi momentum. Note also that the low-frequency boson propagator is peaked at large momenta. But the quasiparticles are renormalized only in a shell of order ω_{ex} close to the Fermi surface. Therefore most of the intermediate scattering states are associated with the conventional band mass. We have, therefore, neglected vertex corrections in this paper.

Scattering by isolated impurities with CEF excitations is usually pair breaking in the s-wave channel⁸ because the dipolar exchange interactions which break the singlet state are stronger than spin-conserving quadrupolar interactions between conduction electrons and CEF states. The s-wave state [where $\Phi(p_z)$ is nodeless] is also not favorable in our case of a periodic lattice of partly localized 5*f* electrons which has dispersive CEF excitations. One can most clearly see this in the following way. In the singlet channel p=-1. Because the kernel $K(\mathbf{q}, i\nu_n)$ is strongly peaked at $q_z = \pi/c$ and $\nu_n = 0$, the gap equation may then be crudely approximated in the following way:

$$\Phi(p_z, i\pi T) \simeq -C\left(p_z, \frac{\pi}{c}\right) \Phi\left(p_z - \frac{\pi}{c}, i\pi T\right), \qquad (34)$$

where $C(p_z, \pi/c)$ is a positive number. Therefore a finite gap $\Phi(p_z, i\pi T)$ must change sign on translation through $Q_z = \pi/c$, i.e., the gap must possess the symmetry $\Phi(p_z - \pi/c) = -\Phi(p_z)$, which excludes an s-wave gap function.

However a non-s-wave superconducting state with nodes may take advantage of the exciton dispersion and become stable. From numerical solutions of the linearized gap equations for our model we find that the instability with the highest T_c is accidentally doubly degenerate. The corresponding gap functions transform as the even- and odd-parity OSP states, $\cos(cp_z)$ and $\sin(cp_z)$. The former is the usual singlet state; the latter is the $S_z=0$ part of the triplet which would appear in a theory with full spin rotational symmetry. We also find an instability in the ESP channel but at a much lower temperature. The corresponding pairing symmetry is $\Phi(p_z) \sim \sin(2cp_z)$. We summarize our results in Table I.

The symmetries of the orbital pair wave functions can be understood in the following way. By comparing our selfenergy equations with those obtained from a four-fermion interaction we see that the effective interaction between quasiparticles in our theory is

$$\langle \gamma_2 \gamma_4 | \hat{V} | \gamma_1 \gamma_3 \rangle = I^2 D^0 \sigma^z_{\gamma_1 \gamma_2} \sigma^z_{\gamma_3 \gamma_4}, \tag{35}$$

or equivalently

$$\hat{V} = I^2 D^0 \hat{\sigma}^z \hat{\sigma}^z. \tag{36}$$

Hence the pairing interaction in some $|\chi\rangle$ channel is

$$V_{\chi}(\mathbf{q}, i\nu_n) = \langle \chi | \hat{\sigma}^z \hat{\sigma}^z | \chi \rangle I^2 D^0(\mathbf{q}, i\nu_n).$$
(37)

We note that our equations go beyond previous calculations^{6,21} in that our formulation includes the full momentum and frequency dependence of the effective interaction $V_{\chi}(\mathbf{q}, i\nu_n)$.

In the OSP channel the static interaction is

$$V(q_z) = -I^2 D^0(q_z) = \frac{I^2 \Delta/2}{\omega(q_z)^2},$$
(38)

which is strongly peaked at $q_z = \pi/c$. The corresponding interaction in real space is therefore attractive when the quasiparticles are separated by a lattice spacing in the *c* direction. Now $\cos(cp_z)$ and $\sin(cp_z)$ are peaked with equal amplitude in real space at z=c. Hence they are equally well suited to take advantage of the attractive part of the interaction and have equal superconducting transition temperatures. The ESP interaction is the negative of the OSP interaction and so it first becomes attractive at z=2c in real space. The only odd-parity wave function which is peaked in real space at this position is $\sin(2cp_z)$.

Ultimately, the degeneracy of the OSP states is a peculiarity of the particular CEF level scheme we adopt for the localized 5f states and of the approximate form of their exciton dispersion which we assume (viz. strongly wave-vector dependent only in the *z* direction). Both OSP states are, in fact, compatible with present experimental evidence. OSP states,





FIG. 1. The dependence of the superconducting transition temperature T_c on the coupling constant g [Eq. (30)]. The band filling n is 0.6.

regardless of the value of total spin angular momentum S, give rise to a reduction in the paramagnetic susceptibility on entering the superconducting state. This reduction comes about because oppositely paired spins have no magnetization. Furthermore, such a reduction in the Pauli susceptibility leads to an upper critical field H_{c2} which, in some cases, can be smaller than that due to the Meissner effect. This effect is called Pauli limiting.²³ A pronounced Knight-shift reduction at Pd sites below T_c (Refs. 13 and 24) and H_{c2} Pauli-limiting²⁵ have both been observed in UPd₂Al₃. Note also that both OSP gap functions have node lines perpendicular to the z direction. In the even-parity case they are located at $p_z = \pm \frac{1}{2}Q_z$, i.e., at the Bragg planes and zone boundaries of the AF Brillouin zone, whereas in the odd-parity case they are located at $p_z=0$. Experimentally, the existence of node lines was inferred from NMR relaxation-rate,^{26,27} specific-heat28 and thermal-conductivity measurements.29 These experiments did not, however, locate the node position along the z direction and so the correct gap symmetry in UPd₂Al₃ presently remains an open problem. To distinguish between the possible nodal gap functions, it is very important to perform field-angle dependent measurements of the specific heat or of the thermal conductivity at low temperature. As proposed in Ref. 30, such measurements may be able to locate the node line in **q** space.

The results of our numerical calculations of the superconducting transition temperature in the ESP and OSP channels are shown in Fig. 1. The superconducting instability occurs first in the OSP channel. We have carried out the calculations with $\omega_{ex}=0.01\epsilon_{\perp}$. This exciton energy corresponds to about 60 K for an electronic bandwidth of 0.5 eV. One therefore obtains a transition temperature in the OSP channel close to the experimental value¹¹ ($T_c=1.8$ K) with a dimensionless coupling constant g/ϵ_{\perp} of about 0.5. At this point we remind the reader that this temperature is in the range of validity of our theory $T_c \ll \omega_{ex} \sim \Delta$.

That contribution to the mass renormalization m^*/m_b (m_b is the band mass) arising from the momentum dependence of the real part of the self-energy is small and so m^*/m_b simply



FIG. 2. The dependence of the mass renormalization m^*/m_b on the coupling constant g [Eq. (30)]. The band filling n is 0.6 as in Fig. 1. m_{min}^* and m_{max}^* are the extremal effective masses over all values of q_z . m_{min}^* occurs at $q_z=0$; m_{max}^* occurs at $q_z=\pi/c$. m^* is practically constant over the Fermi surface for the range of values of g which we consider in this work $(0.5 < g/\epsilon_{\perp} < 4)$.

reduces to the Éliashberg renormalization factor, $Z(\mathbf{p}, i\omega_n)$, which is practically momentum-independent. The weak dependence of $Z(\mathbf{p}, i\omega_n)$ on **p** follows from the combined effect of a strong exciton dispersion along the z axis and a weak electron dispersion in the same direction. Our results for the mass renormalization are shown in Fig. 2. Note that the mass renormalization is approximately linear in $g \sim I^2$ (see Ref. 31). Ab initio calculations of the band masses m_b/m_0 (m_0 is the free electron mass) for the γ ring and β ring of the cylindrical Fermi surface have been carried out by confining two of the three 5f electrons to the uranium ions.⁷ These values, in conjunction with a mass enhancement of m^*/m_b of about 10, yielded values of m^*/m_0 which were in good agreement with experiment.³² We find that such a large value of the mass renormalization corresponds to a value of the coupling constant $(g/\epsilon_{\perp} \approx 2)$ which is roughly a factor of 4 larger than that required to reproduce the transition temperature $(g/\epsilon_{\perp} \approx 0.5)$. One should note that the AF long-range order may act to reduce T_c , an effect which has not been considered here. Therefore the proper value of g/ϵ_{\perp} may indeed be larger than 0.5.

We have also carried out our calculations in the case that the Fermi surface is purely cylindrical (α =0). The results are practically unchanged. This case, however, has the appealing feature that the bracketed factor in the definition of the coupling parameter [see Eq. (30)] is related in the following simple way to the (constant) density of states per spin, $N(\omega)$,

$$g = \frac{I^2 \Delta}{2} N(\mu) \epsilon_{\perp} \frac{1}{\omega_{ex}^2}.$$
 (39)

The dimensionless coupling constant can now be written

$$g/\boldsymbol{\epsilon}_{\perp} = \frac{1}{2} (\lambda_1 \lambda_2)^2 \lambda_3, \tag{40}$$

where the dimensionless constants λ_i are defined by

$$\lambda_1 = IN(\mu), \tag{41}$$

$$\lambda_2 = \Delta/\omega_{ex},\tag{42}$$

$$\lambda_3 = \frac{1}{\Delta N(\mu)}.\tag{43}$$

Of these three parameters, the first, λ_1 , is not presently known for the uranium 5*f* systems. The second, λ_2 , is about unity. The third, λ_3 , is approximately 167, if we make the estimate $N(\mu) \approx 1 \text{ eV}^{-1}$ per cell per spin. The value of the coupling parameter we require to get the experimentally observed transition temperature then leads to the estimate λ_1 ≈ 0.08 . A value of λ_1 which is a factor of 2 larger would yield a mass renormalization in good agreement with experiment, but then T_c would be larger than the experimental value (1.8 K) by roughly a factor of 1.6. As already mentioned, this discrepancy may be alleviated by the effects of AF order. The values for λ_1 obtained here for a dual 5*f* compound are of the same order of magnitude as for 4*f* systems.^{9,31}

IV. CONCLUSIONS AND OUTLOOK

Motivated by experiments on UPd₂Al₃, we have examined a magnetic-exciton model of superconductivity within a strong-coupling retarded framework. The model naturally explains the strong interaction between the collective modes of the localized moments and the heavy conduction electrons which is observed at $\mathbf{Q} = (0, 0, \pi/c)$ in UPd₂Al₃. Solutions of the Éliashberg equations show that the model favors a superconducting instability for the even- or odd-parity OSP states. Each state has line nodes perpendicular to the z direction and total z component of spin $S_z=0$, characteristics which are compatible with measurements of UPd₂Al₃. We find that a superconducting transition temperature and mass renormalization, each in good agreement with their experimental values in UPd₂Al₃, can be obtained using reasonable values of the parameters in the theory. Taken together, these results strengthen the argument in favor of a magnetic-exciton system in UPd₂Al₃.

The evidence for dual f systems in other uranium compounds does not weigh as heavily as it does in the case of UPd₂Al₃. In UPt₃ the number of itinerant f electrons remains controversial. However band-structure calculations based on the assumption that two of the three 5f electrons are localized reproduce the observed de Haas-van Alphen frequencies as well as the anisotropic heavy electron masses in this compound very well.⁷

Recently unconventional superconductivity with split- T_c superconductivity has been discovered in the Pr-based skutterudite $PrOs_4Sb_{12}$.^{33,34} Thermodynamic and transport measurements suggest that the observed heavy-fermion state arises from the interaction of electric quadrupole moments of the CEF-split $4f^2$ states of Pr^{3+} with the conduction electrons.³³ The ground state is probably a singlet and the first excited state is probably a triplet, approximately 0.5 meV higher, with large off-diagonal quadrupolar matrix elements connecting the two states. The largeness of these matrix elements, together with the very small singlet-triplet splitting, strongly suggests an effective mass renormalization via virtual CEF excitations. Finally, experiments reveal that the superconducting order parameter is anisotropic, making this compound another candidate for CEF-exciton-mediated superconductivity, but this time of quadrupolar nature.

UPd₂Al₃ provides a strong motivation for the study of dual *f*-electron models such as the one discussed in this work. We have restricted our study to the case in which all parameters are fixed except the coupling constant. It will be interesting to investigate the dependence of the transition temperature on conduction-band filling and on the form of the exciton dispersion, and to look for a mechanism that can discriminate between the even- and odd-parity superconducting OSP states.

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APPENDIX A: THE ELECTRON-PHONON-LIKE HAMILTONIAN

In this appendix we sketch the derivation of Eq. (5). Our starting point is a set of Holstein-Primakoff-like transformations where the spin operators of the pseudo spin $(S=\frac{1}{2})$ describing the two singlet states are replaced by bosonic operators, *a* and a^{\dagger} , which correspond to the excitation from the singlet ground state to the singlet excited state at energy Δ . This replacement is valid for sufficiently small temperature $(T \ll \Delta)$ when $\langle a^{\dagger}a \rangle \ll 1.^{35}$ We then have

$$S_z = -\frac{1}{2} + a^{\dagger}a, \qquad (A1)$$

$$S^{+} = a S^{-} = a^{\dagger}$$

$$\Rightarrow S_{x} = \frac{1}{2}(a + a^{\dagger}).$$
 (A2)

Inserting these expressions into the second, third, and fourth sums in Eq. (3) and Fourier transforming, we arrive at the following form,

$$\sum_{\mathbf{q}} \left[W_1(\mathbf{q}) a_{\mathbf{q}}^{\dagger} a_{\mathbf{q}} - \frac{W_2(\mathbf{q})}{2} (a_{\mathbf{q}}^{\dagger} a_{-\mathbf{q}}^{\dagger} + a_{\mathbf{q}} a_{-\mathbf{q}}) \right] - \frac{I}{2N} \sum_{\mathbf{p}\mathbf{q}} c_{\mathbf{p}\alpha}^{\dagger} \sigma_{\alpha\beta}^{z} c_{\mathbf{p}+\mathbf{q}\beta} (a_{\mathbf{q}} + a_{-\mathbf{q}}^{\dagger}), \qquad (A3)$$

where

$$W_1(\mathbf{q}) = \Delta - W_2(\mathbf{q}). \tag{A4}$$

The first sum can be diagonalized,³⁵ while at the same time introducing only a factor λ_q in the second sum. To do this we use the Bogoliubov transformation

$$\begin{pmatrix} \alpha_{\mathbf{q}} \\ \alpha^{\dagger}_{-\mathbf{q}} \end{pmatrix} = \begin{pmatrix} u_{\mathbf{q}} & v_{\mathbf{q}} \\ v_{\mathbf{q}} & u_{\mathbf{q}} \end{pmatrix} \begin{pmatrix} a_{\mathbf{q}} \\ a^{\dagger}_{-\mathbf{q}} \end{pmatrix},$$
 (A5)

where $u_q = \cosh(\theta_q)$ and $v_q = \sinh(\theta_q)$. If θ_q is chosen such that

$$-W_2(\mathbf{q})/W_1(\mathbf{q}) = \tanh(2\theta_{\mathbf{q}}), \qquad (A6)$$

then (ignoring constant additive terms) the first sum in Eq. (A3) takes the form,

$$\sum_{\mathbf{q}} \omega_{\mathbf{q}} (\alpha_{\mathbf{q}}^{\dagger} \alpha_{\mathbf{q}} + 1/2), \qquad (A7)$$

where the dispersion $\omega_{\mathbf{q}}$ satisfies

$$\omega_{\mathbf{q}}^2 = W_1^2(\mathbf{q}) - W_2^2(\mathbf{q}).$$
 (A8)

Finally, it is easy to show that

$$a_{\mathbf{q}} + a_{-\mathbf{q}}^{\dagger} = \lambda_{\mathbf{q}} (\alpha_{\mathbf{q}} + \alpha_{-\mathbf{q}}^{\dagger}), \qquad (A9)$$

where

$$\lambda_{\mathbf{q}} = \cosh(\theta_{\mathbf{q}}) - \sinh(\theta_{\mathbf{q}}). \tag{A10}$$

Using Eqs. (A4) and (A6), together with the identity

$$\Delta - 2W_2(\mathbf{q}) = \frac{\omega_{\mathbf{q}}^2}{\Delta},\tag{A11}$$

we can then show that

$$\lambda_{\mathbf{q}}^2 = \frac{\Delta}{\omega_q}.\tag{A12}$$

The interaction term in Eq. (5) follows immediately.

APPENDIX B: THE LINEARIZED GAP EQUATION

The purpose of this appendix is to show how the Isinglike pairing interaction, Eq. (36), leads naturally to two equations for the anomalous self-energy depending on whether the pairing is between spins oriented in the same direction (ESP) or in opposite directions (OSP).

Very recently it has been pointed out³⁶ that a second-order superconducting phase transition can cross over to a firstorder one as one approaches a quantum critical point. There are, however, no indications that UPd_2Al_3 is close to a quantum critical point. Accordingly we assume that the superconducting phase transition is second-order, so that, close to the superconducting transition temperature, the equation for the anomalous self-energy Φ may be linearized,

$$\Phi_{\gamma_{2}\gamma_{4}}(p) = -\frac{T}{N} \sum_{p',\gamma_{1}\gamma_{3}} \langle \gamma_{2}\gamma_{4} | \hat{V}_{p-p'} | \gamma_{1}\gamma_{3} \rangle | G(p') |^{2} \Phi_{\gamma_{1}\gamma_{3}}(p').$$
(B1)

We have adopted a four-vector notation $p = (\mathbf{p}, i\omega_n)$. According to Eq. (36) the matrix elements of the pairing interaction \hat{V} are given by

$$\langle \gamma_2 \gamma_4 | \hat{V} | \gamma_1 \gamma_3 \rangle = I^2 D^0 \langle \gamma_2 \gamma_4 | \hat{\sigma}^z \hat{\sigma}^z | \gamma_1 \gamma_3 \rangle.$$
 (B2)

Now the anomalous self-energy is proportional to the pair wave function. Assuming that we may factorize the latter into a four-momentum-dependent part Φ_p and a spindependent part $|\chi\rangle$, we have

$$\Phi_{\alpha\beta}(p) = \Phi_p \langle \alpha\beta | \chi \rangle. \tag{B3}$$

By inserting Eqs. (B2) and (B3) into Eq. (B1), and using the resolution of the identity,

$$\sum_{\gamma_1\gamma_3} |\gamma_1\gamma_3\rangle \langle \gamma_1\gamma_3| = 1, \qquad (B4)$$

we obtain

$$\Phi_p|\chi\rangle = \hat{\sigma}^z \hat{\sigma}^z |\chi\rangle \frac{T}{N} \sum_{p'} K_{p-p'} |G(p')|^2 \Phi_{p'}, \qquad (B5)$$

where $K = -l^2 D^0$ is the kernel defined in the main text. Taking the scalar product with $\langle \chi |$ leads to the gap equation quoted in the main text, Eq. (11), with the prefactor $p = \langle \chi | \hat{\sigma}^z \hat{\sigma}^z | \chi \rangle$. (This *p* should not be confused with the four momentum.) The Ising-type pairing interaction, Eq. (B2), now allows us to decouple the gap equation into the ESP and OSP channels by virtue of the fact

$$p = \begin{cases} +1 & \text{ESP} \\ -1 & \text{OSP.} \end{cases}$$
(B6)

It is useful to consider the spin structure of the anomalous self-energy in Eq. (B3). The matrix $\langle \alpha\beta | \chi \rangle$ has the form $i\sigma^y$ in the singlet state, S=0. In the triplet state, S=1, its structure is given by $\langle \alpha\beta | \chi \rangle = (i\mathbf{d} \cdot \boldsymbol{\sigma}\sigma^y)_{\alpha\beta}$, where **d** is a complex vector. The ESP states $|1, +1\rangle$ and $|1, -1\rangle$ correspond, respectively, to the **d** vectors $(-\frac{1}{2}, -\frac{1}{2}, 0)$ and $(\frac{1}{2}, -\frac{1}{2}, 0)$, both of which lie in the hexagonal plane. On the other hand, the OSP state $|1, 0\rangle$ corresponds to the **d**-vector $(0, 0, 1/\sqrt{2})$, which is parallel to the *z* axis.

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