Double-stage continuous-discontinuous superconducting phase transition in the Pauli paramagnetic limit of a 3D superconductor: the URu$_2$Si$_2$ case

V. Zhuravlev and T. Maniv

Schulich Faculty of Chemistry, Technion-Israel Institute of Technology, Haifa 32000, Israel

(Dated: February 3, 2009)

The sharp suppression of the de-Haas van-Alphen oscillations observed in the mixed superconducting (SC) state of the heavy fermion compound URu$_2$Si$_2$ is shown to confirm a theoretical prediction of a narrow double-stage SC phase transition, smeared by fluctuations, in a 3D paramagnetically-limited superconductor. The predicted scenario of a second order transition to a nonuniform (FFLO) state followed by a first order transition to a uniform SC state, obtained by using a non-perturbative approach, is also found to be consistent with recent thermal conductivity measurements performed on this material.

The competition between orbital and spin pair-breaking in strongly type-II superconductors in the Pauli paramagnetic limit is known to control the occurrence of discontinuous SC transitions [1,2] at sufficiently low temperatures and high magnetic fields. It was found recently, using perturbation expansion in the SC order parameter [3], that in a clean 3D system, the normal-to-SC phase transitions at low temperatures are of second order, with a SC phase spatially modulated along the field direction [1,3], whereas the transition line from nonuniform-to-uniform SC state was found to be of the first order. This conclusion was reached, however, on the basis of perturbation theory, which might not be valid under the present circumstances due to the following reasons: (1) The jump of the SC order parameter to a finite value at the first order phase transition, and (2) the oscillatory dependence of the quartic and higher order terms in the expansion on the modulation wave number, which makes the utilization of a uniquely defined expression for the SC free energy meaningless within perturbation theory.

The heavy fermion superconductor URu$_2$Si$_2$, whose Fermi surface (FS) may be characterized as 3D [4], possesses characteristic FS parameters which favor strong spin pair breaking. In this material a sharp rise of the thermal conductivity with the decreasing magnetic field just below $H_{c2}$ at low temperatures was reported very recently [5], indicating the existence of a jump in its electronic entropy associated with a first-order phase transition. Furthermore, earlier magneto-oscillations measurements on this material [6] revealed a very sharp damping of the de Haas-van Alpen (dHvA) effect just below $H_{c2}$ which seems to correlate with the anomaly observed in the thermal conductivity.

In this communication we present results of a non-perturbative approach, which establishes the sharp, double-stage transition picture, conjectures in Ref.[3], and argue by means of a detailed theoretical analysis of the experimental dHvA data, that the predicted double-stage transition is realized in URu$_2$Si$_2$. The proposed model is also shown to be consistent with the anomaly in the thermal conductivity data reported in Ref.[5].

We start by writing an expansion of the thermodynamical potential (TP), $\Omega$, in the SC order parameter, $\Delta(\mathbf{r})$, using BCS theory for an isotropic 3D electron gas with the usual $s$-wave electron pairing, as presented in [3]. The use of conventional pairing was made for the sake of simplicity. This is justified in the clean limit considered here since the relevant results have shown in Ref.[3] to be independent of the type of electron pairing. Thus we write:

$$\Omega(\Delta_0) = V \frac{\Delta_0^2}{g_{int}} + \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \Omega_{2n}(\Delta_0), \quad (1)$$

$$\Omega_{2n} = \int d^3 \{r\} \Gamma_{2n}(\{r\}, \Delta_0) \tilde{K}_{2n}(\{r\})$$

where $g_{int}$ is the effective BCS coupling constant, $V$ is the volume:

$$\Gamma_{2n}(\{r\}, \Delta_0) = g^\ast(r_1, r_2) g(r_2, r_3) \cdots g^\ast(r_{2n-1}, r_{2n}) \times g(r_{2n}, r_1) \Delta(r_1) \Delta^\ast(r_2) \cdots \Delta(r_{2n-1}) \Delta^\ast(r_{2n}) \quad (2)$$

and:

$$\tilde{K}_{2n}(\{r\}) = k_B T \sum_{\nu} \overline{G}_{01}(r_1, r_2, \omega, \nu) \overline{G}_{01}(r_2, r_3, \omega, \nu) \cdots \overline{G}_{01}(r_{2n-1}, r_{2n}, \omega, \nu) \overline{G}_{01}(r_{2n}, r_1, \omega, \nu) \quad (3)$$

Here $\Delta_0^2 = V^{-1} \int d^3 r_i |\Delta(r_i)|^2$, and $\{r\} = \{r_1, \ldots, r_{2n}\}$ denotes the entire set of position vectors for a cluster consisting of $n$ electron pairs. Note that for convenience we incorporated the gauge factors, $g(r_1, r_{i+1})$, of the Green’s functions, $G_{01}(r_i, r_{i+1}, \omega, \nu)$, for a free electron in a uniform magnetic field, into the vertex part, $\Gamma_{2n}$, so that the effective kernel $\tilde{K}_{2n}$ is given in Eq. (3) by a product of the gauge invariant Green’s functions, $\overline{G}_{01}(r_i, r_{i+1}, \omega, \nu)$. A useful expression for such a Green’s function for a positive Matsubara frequency, $\nu \geq 0$, can be written as:

$$\overline{G}_{01}(r_1, r_2, \omega) = \frac{1}{2\pi a_H^2 \omega c} \int \frac{dk_z}{2\pi} e^{ik_z(z_2-z_1)} e^{-p_z^2/4} \int_0^\infty d\tau e^{i\tau [n_F + g - x^2 + iz_2]} (1 - e^{-i\tau})^{-1} \exp \left( -\frac{\rho^2 e^{-i\tau}}{2(1 - e^{-i\tau})} \right),$$

$$\Gamma_{2n}(\{r\}) = g^\ast(r_1, r_2) g(r_2, r_3) \cdots g^\ast(r_{2n-1}, r_{2n}) \times g(r_{2n}, r_1) \Delta(r_1) \Delta^\ast(r_2) \cdots \Delta(r_{2n-1}) \Delta^\ast(r_{2n}) \quad (2)$$

and:

$$\tilde{K}_{2n}(\{r\}) = k_B T \sum_{\nu} \overline{G}_{01}(r_1, r_2, \omega, \nu) \overline{G}_{01}(r_2, r_3, \omega, \nu) \cdots \overline{G}_{01}(r_{2n-1}, r_{2n}, \omega, \nu) \overline{G}_{01}(r_{2n}, r_1, \omega, \nu) \quad (3)$$

Here $\Delta_0^2 = V^{-1} \int d^3 r_i |\Delta(r_i)|^2$, and $\{r\} = \{r_1, \ldots, r_{2n}\}$ denotes the entire set of position vectors for a cluster consisting of $n$ electron pairs. Note that for convenience we incorporated the gauge factors, $g(r_1, r_{i+1})$, of the Green’s functions, $G_{01}(r_i, r_{i+1}, \omega, \nu)$, for a free electron in a uniform magnetic field, into the vertex part, $\Gamma_{2n}$, so that the effective kernel $\tilde{K}_{2n}$ is given in Eq. (3) by a product of the gauge invariant Green’s functions, $\overline{G}_{01}(r_i, r_{i+1}, \omega, \nu)$. A useful expression for such a Green’s function for a positive Matsubara frequency, $\nu \geq 0$, can be written as:

$$\overline{G}_{01}(r_1, r_2, \omega) = \frac{1}{2\pi a_H^2 \omega c} \int \frac{dk_z}{2\pi} e^{ik_z(z_2-z_1)} e^{-p_z^2/4} \int_0^\infty d\tau e^{i\tau [n_F + g - x^2 + iz_2]} (1 - e^{-i\tau})^{-1} \exp \left( -\frac{\rho^2 e^{-i\tau}}{2(1 - e^{-i\tau})} \right),$$

$$\Gamma_{2n}(\{r\}) = g^\ast(r_1, r_2) g(r_2, r_3) \cdots g^\ast(r_{2n-1}, r_{2n}) \times g(r_{2n}, r_1) \Delta(r_1) \Delta^\ast(r_2) \cdots \Delta(r_{2n-1}) \Delta^\ast(r_{2n}) \quad (2)$$

and:

$$\tilde{K}_{2n}(\{r\}) = k_B T \sum_{\nu} \overline{G}_{01}(r_1, r_2, \omega, \nu) \overline{G}_{01}(r_2, r_3, \omega, \nu) \cdots \overline{G}_{01}(r_{2n-1}, r_{2n}, \omega, \nu) \overline{G}_{01}(r_{2n}, r_1, \omega, \nu) \quad (3)$$

Here $\Delta_0^2 = V^{-1} \int d^3 r_i |\Delta(r_i)|^2$, and $\{r\} = \{r_1, \ldots, r_{2n}\}$ denotes the entire set of position vectors for a cluster consisting of $n$ electron pairs. Note that for convenience we incorporated the gauge factors, $g(r_1, r_{i+1})$, of the Green’s functions, $G_{01}(r_i, r_{i+1}, \omega, \nu)$, for a free electron in a uniform magnetic field, into the vertex part, $\Gamma_{2n}$, so that the effective kernel $\tilde{K}_{2n}$ is given in Eq. (3) by a product of the gauge invariant Green’s functions, $\overline{G}_{01}(r_i, r_{i+1}, \omega, \nu)$. A useful expression for such a Green’s function for a positive Matsubara frequency, $\nu \geq 0$, can be written as:

$$\overline{G}_{01}(r_1, r_2, \omega) = \frac{1}{2\pi a_H^2 \omega c} \int \frac{dk_z}{2\pi} e^{ik_z(z_2-z_1)} e^{-p_z^2/4} \int_0^\infty d\tau e^{i\tau [n_F + g - x^2 + iz_2]} (1 - e^{-i\tau})^{-1} \exp \left( -\frac{\rho^2 e^{-i\tau}}{2(1 - e^{-i\tau})} \right),$$
where $\rho_{1,2} \equiv r_{1,2} - r_{1,1}$, with $r_{1,1}$, $r_{1,2}$ the projections of the initial and final electron position vectors, respectively, on the $(x-y)$ plane perpendicular to the magnetic field. This expression is obtained after summation over the Landau level (LL) index $n = 0, 1, \ldots$, of the single-particle energy, $\varepsilon_{n1}/\omega_c = n + k_z^2/2m + i\omega_c$, for a spin up (or down) electron in a magnetic field $H = H_2^x$, with a cyclotron frequency $\omega_c = eH/m^*c$, Zeeman spin energy $\mp eH/m^*c$, and g-factor $g = m^*/m_0$, with $m^*$, and $m_0$ the effective mass and free electron mass respectively. Here $k_2^2 = H^2 k^2/2m^*\omega_c$, $n_F = \pi/\omega_c$, $\mu$, the chemical potential ($\approx E_F$ - Fermi energy), and $\omega_c = \omega_F/\omega_v$ with $\omega_v = \pi k_B T/(2\nu + 1)/\hbar$. For negative Matsubara frequencies, $\nu < 0$, a similar expression can be derived by replacing $\tau$ with $-\tau$. In what follows we will express space coordinates and momenta in units of $a_H = \sqrt{\hbar/eH}$ and $a_H^{-1}$ respectively. The SC order parameter is assumed to take the form, $\Delta(r, z) = \Delta_{\text{max}} e^{i q z} \varphi_0(x, y)$, where $\Delta^2_{\text{max}} = (2\pi \nu)^{1/2} \Delta_0^2$ and $\varphi_0(x, y)$ describes (in the symmetric gauge) an hexagonal vortex lattice with inter-vortex distance, $a_x = \sqrt{\pi \nu}$. Here $e^{i q z}$ is a Fulde-Ferrel (FF) modulation function along the magnetic field direction, controlled by the wave-number, $q$.

The vertex part $F_{2\nu}(\{r\}, \Delta_0)$, Eq. (2), is a violently oscillating function of the lateral relative electronic coordinates, which interferes strongly with the oscillatory electronic kernel $K_{2\nu}(\{r\})$, Eq. (3). Multiple integration over these coordinates yields gross cancellations except near stationary configurations, which restrict all 2n electronic position vectors to a relative proximity region of size of a magnetic length $L_\phi$. Other contributions to this integral, arising from non-stationary, separately paired configurations, become increasingly important in random vortex lattices where the phase coherence responsible for the constructively interfering configurations breakdown. Whereas the small oscillatory (high harmonic in $1/H$) part of the TP is strongly influenced by these non-local contributions, their influence on the much larger non-oscillatory (zero harmonic in $1/H$) component is not important (see Ref. [5]). The great advantage of using the local approximation in Eq. (11) is in its factorization with respect to the relative coordinates and its apparent independence of the center of mass coordinates, which enable us rewriting the integrand in Eq. (11) as a separable product of effective single electron Green’s functions. The corresponding $n$-th order term, $\Omega_n$, can be thus written as a $3D$ integral over the center of mass momentum in an effective two-particle Green’s function, raised to the $n$-th power, by performing an appropriate Fourier-transformation, namely: $\Omega_n(\Delta_{\text{max}}) = V \frac{\alpha_{n1}}{(2\pi \nu)^3} \frac{\Delta^2_{\text{max}}}{(\hbar \omega_c)^2} I_{2n}$, where $\Delta^2_{\text{max}} = \Delta_{\text{max}}^2 (\hbar \omega_c)^2$, and:

$$I_{2n} = \sum_{\nu > 0} \int \frac{d^2 k dk}{(2\pi \nu)^3} \left[ \Phi_{\nu}(k, k_z | g, q) \Phi^*_{\nu}(k, k_z | -g, -q) \right]^{n} + \ldots$$

with: $\Phi_{\nu}(k, k_z | g, q) = \int_0^{\infty} d\tau e^{-\tau} [\omega_c i\nu g_{k, k_z} - \frac{1}{2}(1 + \tau + \tau^2)] |k|^2$.

This resulting perturbation series can be easily summed to all orders, provided the reduction pre-factor $1/\sqrt{n}$, arising from the overlap integral of $n$ lowest LL orbitals is represented as a Gaussian integral: $\frac{2}{\sqrt{\pi}} \int_0^{\infty} d\frac{\mu}{\sigma} \exp\left(-\left(n\mu^2\right)\right)$. In the quasi-classical limit $\tau < 1$, the Gaussian approximation $e^{-\frac{1}{4} \left(1 + \tau + \tau^2\right)} |k|^2 \approx e^{-\frac{1}{4} \tau^2 |k|^2}$, accounts for the diamagnetic pair-breaking whereas all quantum corrections (including quantum magnetic oscillations), which arise near the lattice points $\tau = 2\pi l$, $l \neq 0$, are neglected.

It is convenient to normalize all energies by $\pi k_B T_c$, and $T_c$, is the transition temperature $H = 0$, so that: $\Delta_{\text{max}}(H = 0) = \Delta_{\text{max}}(T_c) = \frac{\mu}{\pi k_B T_c}$, $\tau = \frac{\hbar}{k_B T_c}$, $\pi k_B T_c$, $\tau = \frac{\hbar}{k_B T_c}$, $\pi k_B T_c$, and $\gamma = \frac{a^2 h e}{\pi k_B T_c}$, where $\omega_{c20} = \pi k_B T_c/m^*c$, and $H_{c20}$ is the theoretical upper critical field at $T = 0$ (in the absence of spin splitting). Performing the integration over $\tau$, the resulting expression for TP can be written in the form:

$$\Omega(\pi k_B T_c) = \frac{\Delta^2_{\text{max}}}{\lambda} - \frac{t}{\pi} \frac{1}{\lambda^2} Re \sum_{\nu > 0} \int \left\{ \int d\kappa d\kappa_z \ln \left(1 + e^{-\nu^2} \Delta_{\text{max}}(\nu g, \nu q) \Phi^*_{\nu}(k, k_z) \right) \right\}_0 \Phi_{\nu}(k, k_z | g, q)$$

where $k = \sqrt{k_x^2 + k_y^2 + k_z^2}$, $\kappa = \sqrt{k_x^2 + k_y^2 + k_z^2}$, $\nu = \phi_{k, k_z} + \phi_{k, k_z}$, $\omega_{c20} = \pi k_B T_c/m^*c$, and $H_{c20}$ is the zero temperature coherence length.

In the limit of zero spin splitting and in the absence of FF modulation ($g = q = 0$) the effective pairing parameter, $\chi_{\nu}(k, k_z) \equiv \Delta_{\text{max}}(\nu g, \nu q)$, is always real and positive. Under these circumstances the general form of $\Omega(\pi k_B T_c)$ at $H < H_{c20}$, as expressed in Eq. (4), possesses the single minimum structure characterizing the usual GL theory. For $g \neq 0$, $\chi_{\nu}(k, k_z)$ can become complex (a feature that can be "healed" by the presence of the FF modulation wavenumber $q$), so that the general form of $\Omega(\pi k_B T_c)$ may show a maximum at small $\Delta_{\text{max}}$ which is followed by a minimum at large $\Delta_{\text{max}}$. The initial maximum reflects the competition between the increasing spin paramagnetic energy and decreasing SC pair-correlation energy as the number of spin-singlet Cooper-pairs is increased.

Typical results of the calculated TP using Eq. (4) for non-zero spin-splitting are shown in Fig. 1. As discussed
and the usual single minimum picture is restored (see the dashed curves in Fig. 1). Thus, instead of the "expected" first-order transition to a uniform SC state one finds a second-order phase transition to a nonuniform (FF) SC state.

However, due to its compensation effect, the FF modulation significantly reduces the equilibrium SC free energy with respect to its uniform counterpart (compare the dashed curves to the corresponding solid ones). As a result, the field range of stability of the modulated phase is quite small. Thus, by slightly reducing the field below the second order normal-to-SC transition the $q = 0$ state becomes energetically more favorable and the system transforms from the nonuniform to a uniform SC state via a first-order phase transition. One should note that the second order perturbation theory with $\Omega / \Xi_0 = \alpha (q) \Delta q_{\text{max}} + \frac{1}{2} \beta (q) \Delta q_{\text{max}}^2$, is quantitatively correct only for small values of the order parameter, namely for $\Delta q_{\text{max}} \lesssim 0.1$, and, therefore, cannot be applied to the first order transition.

Fig. 2 shows the calculated self-consistent $\Delta q_{\text{max}}^2$ (which is optimized with respect to $q$) as a function of magnetic field at a temperature well below the tri-critical temperature and for characteristic parameters corresponding to URu$_2$Si$_2$. The initial build-up of the SC order parameter in a narrow region following the second-order phase transition and the pronounced jump at the next (first order) transition are apparent. The overall width of the two-stage transition ($\Delta b / b \approx 0.06$) is in good agreement with the width of the sharp structure observed experimentally in the thermal transport measurements [7].

Fig. 3 exhibits the result of a detailed fitting procedure of our calculation to the experimental data of the dHvA oscillations observed by Ohkuni et. al [3] in the mixed state of URu$_2$Si$_2$. The data shows a very sharp reduction in the amplitude of the dHvA oscillation, $A_{\text{SC}}$, just below $H = 2.78$ T, similar to the step-like structure observed in the thermal conductivity measurements [7]. For fitting the measured relative signal, $A_{\text{SC}} / A_n$ (where $A_n$ being the theoretical dHvA amplitude as extrapolated from the normal state) in the region above the sharp damping interval, we exploit the fluctuating vortex lattice model described in Refs. [9, 10] and write: $\ln (A_{\text{SC}} / A_n) = -\frac{\pi^2}{4 \hbar c \sqrt{E F / \Delta_{\text{max}}}} \langle \Delta^2 \rangle$, where $\langle \Delta^2 \rangle$ is the mean-square order parameter for the modulated FF state in the vicinity of the second-order phase transition, i.e.: $\langle \Delta^2 \rangle = \frac{\pi^2}{2} \Delta_{\text{max}}^2 \left( 1 + \sqrt{1 + \nu^2 / x^2} \right)$, with $x = \frac{\alpha}{\sqrt{2}} \left( \frac{\hbar c v_0}{k_B T} \Delta_{\text{max}} \right)$ and $\nu = 0.51$ for a 3D system (see Ref. [10]). Here $\beta$ is the coefficient of the quartic term obtained in the expansion of $\Delta_{\text{max}}$ in $\Delta_{\text{max}}$ near the SC transition, and $v_0 = \pi^2 a^2 H_c / k_F$. The selected value of $E_F$ has been determined from the experimentally observed, dominant dHvA frequency, $F_0 \approx 10^5$ T, corresponding to the nearly spherical band 17-hole Fermi
FIG. 3: (color online) Logarithm of the dHvA amplitude ratio $A_S/A_n$ reported in Ref.[6] (full circles) and the values of $-\chi \langle \Delta^2 \rangle$, $\chi = \frac{\alpha^n}{\hbar c_0 \sqrt{K_p h_c}}$ as obtained from our calculation near the second order phase transition (solid line), as functions of $1/H$. The dashed broken straight line is our mean-field result for $-\chi \Delta^2_{\text{max}}$ around the first-order transition. The dotted line represents the extrapolation of $-\chi \Delta^2_{\text{max}}$ obtained in our calculation is nearly independent of the various parameters involved, provided the temperature $t$ is well below the tri-critical point $t_{\text{tric}}$.

In conclusion, using a non-perturbative approach, we have firmly established our early conjecture concerning the SC transition in a 3D strongly type-II superconductor in the paramagnetic limit, and show that the dHvA effect observed in the mixed SC state of URu$_2$Si$_2$[6] provides a clear experimental evidence for the double-stage nature of this transition, which is smeared by significant SC fluctuations effect. This finding is consistent with the interpretation of a first-order phase transition given in Ref.[7] to the step-like structure observed in the thermal transport data of this material. We note that the unusual sign of the observed jump in the thermal conductivity, which could be due to some peculiar quasi-particle scattering mechanism [11], is irrelevant to our main argument, which associates this jump, irrespective of its direction, to the jump of the SC order parameter at the predicted first-order transition.

We thank J. Wosnitza, B. Bergk and Y. Kasahara for valuable discussions. This research was supported by the Israel Science Foundation founded by the Academy of Sciences and Humanities, by Posansky Research fund in superconductivity, and by EuroMagNET under the EU contract RI3-CT-2004-506239.

[1] G. Sarma, J. Phys. Chem. Solids 24, 1029 (1963).
[2] K. Maki and T. Tsuneto, Prog. Theor. Phys. 31, 945 (1964).
[3] T. Maniv and V. Zhuravlev, Phys. Rev. B 77, 134511 (2008).
[4] P. Fulde and R.A. Ferrell, Phys. Rev. 135, A550, (1964).
[5] A.I. Larkin and Yu.N. Ovchinnikov, Zh. Eksp. Teor. Fiz. 47, 1136 (1964) [Sov. Phys. JETP 20, 762 (1965)]
[6] H. Ohkuni, Y. Inada, Y. Tokiwa, K. Sakurai, R. Settai, T. Honma, Y. Haga, E. Yamamoto, Y. Onuki, H. Yamagami, S. Takahashi and T. Yanagisawa, Phil. Mag. B 79, 1045 (1999).
[7] Y. Kasahara et al., Phys. Rev. Lett. 99, 116402 (2007).
[8] V. Zhuravlev, T. Maniv, I. D. Vagner, and P. Wyder, Phys. Rev. B 56, 14693 (1997).
[9] T. Maniv, V. Zhuravlev, I. D. Vagner, and P. Wyder, Rev. Mod. Phys. 73, 867.
[10] T. Maniv, V. Zhuravlev, J. Wosnitza, O. Ignatchik, B. Bergk, and P.C. Canfield, Phys. Rev. B 73, 134521-7 (2006).
[11] Hiroto Adachi and M. Sigrist, arXiv [cond-mat.supr-con]: 0710.3110