

## Fixed- $N$ Superconductivity: The Crossover from the Bulk to the Few-Electron Limit

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We present a truly canonical theory of superconductivity in ultrasmall metallic grains by variationally optimizing fixed- $N$  projected BCS wave functions, which yields the first full description of the *entire crossover* from the bulk BCS regime (mean level spacing  $d \ll$  bulk gap  $\tilde{\Delta}$ ) to the “fluctuation-dominated” few-electron regime ( $d \gg \tilde{\Delta}$ ). A wave-function analysis shows in detail how the BCS limit is recovered for  $d \ll \tilde{\Delta}$ , and how for  $d \gg \tilde{\Delta}$  pairing correlations become delocalized in energy space. An earlier grand-canonical prediction for an observable parity effect in the spectral gaps is found to survive the fixed- $N$  projection. [S0031-9007(98)07675-3]

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In the early days of BCS theory, its use of essentially grand-canonical (g.c.) wave functions was viewed as one of its most innovative, if not perplexing, features: the variational BCS ansatz for the ground state is a superposition of states with different electron numbers, although BCS [1] themselves had emphasized that the true ground state of an isolated superconductor must be a state of definite electron number. That this ansatz was nevertheless rapidly accepted and tremendously successful had two reasons: first, calculational convenience—determining the variational parameters is incomparably much simpler in a g.c. framework, where the particle number is fixed only on the average, than in a canonical one, where a further projection to fixed electron number is required; and second, it becomes exact in the thermodynamic limit—fixed- $N$  projections yield corrections to the BCS ground state energy per electron that are only of order  $N^{-1}$ , as shown, e.g., by Anderson [2] and Mühlischlegel [3].

Recently, however, a more detailed examination of the range of validity of BCS’s g.c. treatment has become necessary, in light of the measurements by Ralph, Black, and Tinkham (RBT) [4] of the discrete electronic spectrum of an individual ultrasmall superconducting grain: it had a charging energy so large ( $E_C \gg \tilde{\Delta}$ ) that electron number fluctuations are strongly suppressed, calling for a canonical description, and the number of electrons  $N$  within the Debye frequency cutoff  $\omega_D$  from the Fermi energy  $\varepsilon_F$  was only of order  $10^2$ ; hence, differences between canonical and g.c. treatments might become important. Moreover, its mean level spacing  $d \propto N^{-1}$  was comparable to the bulk gap  $\tilde{\Delta}$ ; hence, it lies right in the *crossover regime* between the “fluctuation-dominated” (f.d.) few-electron regime ( $d \gg \tilde{\Delta}$ ) and the bulk BCS regime ( $d \ll \tilde{\Delta}$ ), which could not be treated satisfactorily in any of the recent theoretical papers inspired by these experiments: the results of [5–9], including the predictions of parity effects, were obtained in a g.c. framework; and Mastellone, Falci, and Fazio’s (MFF) [10] fixed- $N$  exact numerical diagonalization study, the first detailed analysis of the fluctuation-dominated regime, was limited to  $N \leq 25$ .

In this Letter we achieve the *first canonical description of the full crossover*. We explicitly project the BCS ansatz to fixed  $N$  (for  $N \leq 600$ ) before variationally optimizing it, adapting an approach developed by Dietrich, Mang, and Pradal [11] for shell-model nuclei with pairing interactions to the case of ultrasmall grains. This projected BCS (PBCS) approach enables us (i) to significantly improve previous g.c. upper bounds on ground state energies [5–8], (ii) to check that a previous grand-canonical prediction [8] for an *observable* parity effect in the spectral gaps survives the fixed- $N$  projection, (iii) to find in the crossover regime a remnant of the “breakdown of superconductivity” found in g.c. studies, at which the condensation energy changes from being extensive to practically intensive, and (iv) to study this change by an *explicit wave-function analysis*, which shows in detail how the BCS limit is recovered for  $d \ll \tilde{\Delta}$ , and how for  $d \gg \tilde{\Delta}$  pairing correlations become delocalized in energy space.

*The model.*—We model the superconducting grain by a reduced BCS Hamiltonian which has been used before to describe small superconducting grains [6–9] (it was phenomenologically successful for  $d \lesssim \tilde{\Delta}$  [7,8], but probably is unrealistically simple for  $d \gg \tilde{\Delta}$ , for which it should rather be viewed as a toy model):

$$H = \sum_{j=0,\sigma}^{N-1} \varepsilon_j c_{j\sigma}^\dagger c_{j\sigma} - \lambda d \sum_{j,j'=0}^{N-1} c_{j+}^\dagger c_{j-}^\dagger c_{j'-} c_{j'+}. \quad (1)$$

The  $c_{j\pm}^\dagger$  create electrons in free time-reversed single-particle-in-a-box states  $|j, \pm\rangle$ , with discrete, uniformly spaced, degenerate eigenenergies  $\varepsilon_j = jd + \varepsilon_0$ . The interaction scatters only time-reversed pairs of electrons within  $\omega_D$  of  $\varepsilon_F$ . Its dimensionless strength  $\lambda$  is related to the two material parameters  $\tilde{\Delta}$  and  $\omega_D$  via the bulk gap equation  $\sinh 1/\lambda = \omega_D/\tilde{\Delta}$ . We chose  $\lambda = 0.22$ , close to that of Al [8]. The level spacing  $d$  determines the number  $N = 2\omega_D/d$  of levels, taken symmetrically around  $\varepsilon_F$ , within the cutoff; electrons outside the cutoff remain unaffected by the interaction and are thus neglected throughout.

*Projected variational method.*—We construct variational ground states for  $H$  by projecting BCS-type wave

functions onto a fixed electron number  $N = 2n_0 + b$  [11], where  $n_0$  and  $b$  are the number of electron pairs and unpaired electrons within the cutoff, respectively. Considering  $b = 0$  first, we take

$$|0\rangle = C \int_0^{2\pi} d\phi e^{-i\phi n_0} \prod_{j=0}^{N-1} (u_j + e^{i\phi} v_j c_{j+}^\dagger c_{j-}^\dagger) |\text{vac}\rangle, \quad (2)$$

where  $|\text{vac}\rangle$  is the vacuum state. Both  $v_j$ , the amplitude to find a pair of electrons in the levels  $|j, \pm\rangle$ , and  $u_j$ , the amplitude for the levels being empty, can be chosen real [11] and obey  $u_j^2 + v_j^2 = 1$ . The integral over  $\phi$  performs the projection onto the fixed electron pair number  $n_0$ , and  $C$  is a normalization constant ensuring  $\langle 0|0\rangle = 1$ .

Doing the integral analytically yields a sum over  $\binom{2n_0}{n_0}$  terms [all products in (2) that contain exactly  $n_0$  factors of  $v_j c_{j+}^\dagger c_{j-}^\dagger$ ], which is forbiddingly unhandy for

$$\Lambda_j \equiv \sum_k \left( \varepsilon_j - \frac{\lambda d}{2} \right) v_k^2 \left[ \frac{R_2^{jk} - R_1^{jk}}{R_0} - \frac{R_1^k R_1^j - R_0^j}{R_0} \right] - \frac{\lambda d}{2} \sum_{k,\ell} u_k v_k u_\ell v_\ell \left[ \frac{R_2^{jk\ell} - R_1^{jk\ell}}{R_0} - \frac{R_1^{k\ell} R_1^j - R_0^j}{R_0} \right].$$

We obtain an upper bound on the ground state energy and a set of  $v_j$ 's, i.e., an approximate wave function, by solving these equations numerically. To this end, we use a formula of Ma and Rasmussen [12] to express any  $R_n^{j_1 \dots j_N}$  in terms of  $R_0$  and all  $R_0^j$ 's, and evaluate the latter integrals using fast Fourier transform routines.

Next consider states with  $b$  unpaired electrons, e.g., states with odd number parity or excited states: Unpaired electrons are "inert" because the particular form of the interaction involves only electron *pairs*. Thus the Hilbert subspace with  $b$  specific levels occupied by unpaired electrons, i.e., levels "blocked" to pair scattering [7,13], is closed under the action of  $H$ , allowing us to calculate the energy, say  $\mathcal{E}_b$ , of its ground state  $|b\rangle$  by the variational method also. To minimize the kinetic energy of the unpaired electrons in  $|b\rangle$  we choose the  $b$  singly occupied levels,  $j \in B$ , to be those closest to the Fermi surface [8]. Our variational ansatz for  $|b\rangle$  then differs from  $|0\rangle$  only in that  $\prod_j$  is replaced by  $(\prod_{j \in B} c_{j+}^\dagger) \prod_{j \notin B}$ . Thus in all products and sums over  $j$  above, the blocked levels are excluded (the  $u_j$  and  $v_j$  are not defined for  $j \in B$ ) and the total energy  $\mathcal{E}_b$  has an extra kinetic term  $\sum_{j \in B} \varepsilon_j$ .

In the limit  $d \rightarrow 0$  at fixed  $n_0 d$ , the PBCS theory reduces to the g.c. BCS theory of Ref. [7] (proving that the latter's  $N$  fluctuations become negligible in this limit): The projection integrals can then be approximated by their saddle point values [11]; since  $\phi = 0$  at the saddle, the  $R$ 's used here are all equal, thus  $\Lambda_j$  vanishes, the variational equations decouple and reduce to the BCS gap equation, and the saddle point condition fixes the *mean* number of electrons to be  $2n_0 + b$ . To check the opposite limit of  $d \gg \tilde{\Delta}$  where  $n_0$  becomes small, i.e., the f.d. regime, we compared our PBCS results for  $\mathcal{E}_0$  and  $\mathcal{E}_1$  with MFF's

any reasonable  $n_0$ . Therefore we follow Ref. [11] and evaluate all integrals numerically instead. Introducing the following shorthand for a general *projection integral*,

$$R_n^{j_1 \dots j_N} \equiv \int_0^{2\pi} \frac{d\phi}{2\pi} e^{-i(n_0 - n)\phi} \prod_{j \neq j_1 \dots j_N} (u_j^2 + e^{i\phi} v_j^2),$$

the expectation value  $\mathcal{E}_0 = \langle 0|H|0\rangle$  can be expressed as

$$\mathcal{E}_0 = \sum_j (2\varepsilon_j - \lambda d) v_j^2 \frac{R_1^j}{R_0} - \lambda d \sum_{j,k} u_j v_j u_k v_k \frac{R_1^{jk}}{R_0}.$$

Minimization with respect to the variational parameters  $v_j$  leads to a set of  $2n_0$  coupled equations,

$$2(\hat{\varepsilon}_j + \Lambda_j) u_j v_j = \Delta_j (u_j^2 - v_j^2), \quad (3)$$

where the quantities  $\hat{\varepsilon}_j$ ,  $\Lambda_j$ , and  $\Delta_j$  are defined by

$$\hat{\varepsilon}_j \equiv (\varepsilon_j - \lambda d/2) \frac{R_1^j}{R_0}, \quad \Delta_j \equiv \lambda d \sum_k u_k v_k \frac{R_1^{jk}}{R_0},$$

exact results [10], finding agreement to within 1% for  $n_0 \leq 12$ . This shows that "superconducting fluctuations" (as pairing correlations are traditionally called when, as in this regime, the g.c. pairing parameter vanishes [6]) are treated adequately in the PBCS approach. Because it works so well for  $d \ll \tilde{\Delta}$  and  $d \gg \tilde{\Delta}$ , it seems reasonable to trust it in the crossover regime  $d \approx \tilde{\Delta}$  also, though here, lacking any exact results for comparison, we cannot quantify its errors.

*Ground state energies.*—Figure 1(a) shows the ground state condensation energies  $E_b = \mathcal{E}_b - \langle F_b | H | F_b \rangle$  for even and odd grains ( $b = 0$  and  $1$ , respectively), which is measured relative to the energy of the respective uncorrelated Fermi sea ( $|F_0\rangle = \prod_{j < n_0} c_{j+}^\dagger c_{j-}^\dagger |\text{vac}\rangle$  or  $|F_1\rangle = c_{n_0+}^\dagger |F_0\rangle$ ), calculated for  $N \leq 600$  using both the canonical ( $E_b^C$ ) and g.c. ( $E_b^{\text{GC}}$ ) [6,7] approaches. The g.c. curves suggest a breakdown of superconductivity [6,7] for large  $d$ , in that  $E_b^{\text{GC}} = 0$  above some critical  $b$ -dependent level spacing  $d_b^{\text{GC}}$ . In contrast, the  $E_b^C$ 's are (i) significantly lower than the  $E_b^{\text{GC}}$ 's, thus the projection much improves the variational ansatz, and (ii) negative for *all*  $d$ , which shows that the system can *always* gain energy by allowing pairing correlations, even for arbitrarily large  $d$ . As anticipated in [8], the breakdown of superconductivity is evidently not as complete in the canonical as in the g.c. case. Nevertheless, some remnant of it does survive in  $E_b^C$ , since its behavior also changes markedly at a  $b$ (and  $\lambda$ )-dependent characteristic level spacing  $d_b^C$  ( $< d_b^{\text{GC}}$ ): it marks the end of bulk BCS-like behavior for  $d < d_b^C$ , where  $E_b^C$  is *extensive* ( $\sim 1/d$ ), and the start of a f.d. plateau for  $d > d_b^C$ , where  $E_b^C$  is practically *intensive* (almost  $d$  independent) [14]. The standard heuristic interpretation [15] of the bulk BCS limit  $-\tilde{\Delta}^2/2d$  (which is indeed reached

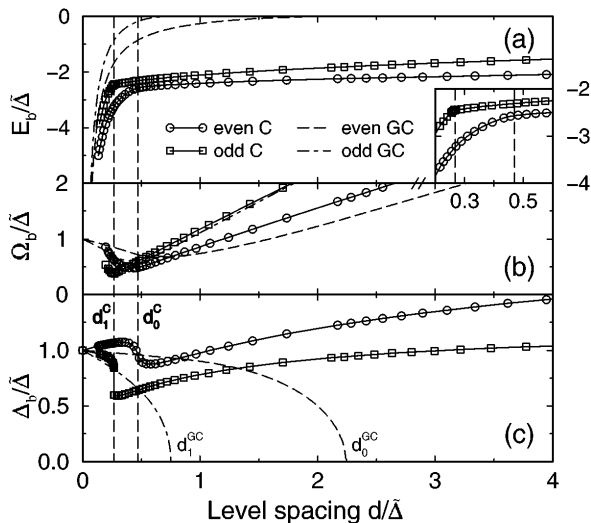


FIG. 1. (a) The ground state condensation energies  $E_b$ , (b) the spectral gaps  $\Omega_b = \mathcal{E}_{b+2} - \mathcal{E}_b$ , and (c) the pairing parameters  $\Delta_b$ , for even and odd systems ( $b = 0, 1$ ), calculated canonically (C) and grand canonically (GC) as functions of  $d/\tilde{\Delta} = 2 \sinh(1/\lambda)/N$ . The inset shows a blowup of the region around the characteristic level spacings  $d_0^C = 0.5\tilde{\Delta}$  and  $d_1^C = 0.25\tilde{\Delta}$  (indicated by vertical lines in all subfigures). The  $d_b^C$  (a) mark a change in behavior of  $E_b$  from  $\sim 1/d$  to being almost  $d$  independent, and roughly coincide with (b) the minima in  $\Omega_b$ , and (c) the position of the abrupt drops in  $\Delta_b$ .

by  $E_b^C$  for  $d \rightarrow 0$ ) hinges on the scale  $\tilde{\Delta}$ : the number of levels strongly affected by pairing is roughly  $\tilde{\Delta}/d$  (those within  $\tilde{\Delta}$  of  $\varepsilon_F$ ), with an average energy gain per level of  $-\tilde{\Delta}/2$ . To analogously interpret the  $d$  independence of  $E_b^C$  in the f.d. regime, we argue that *the scale  $\tilde{\Delta}$  loses its significance*—fluctuations affect *all*  $n_0 = \omega_D/d$  unblocked levels within  $\omega_D$  of  $\varepsilon_F$  (this is made more precise below), and the energy gain per level is proportional to a renormalized coupling  $-\tilde{\lambda}d$  (corresponding to the  $1/N$  correction of [2,3] to the g.c. BCS result). The inset of Fig. 1(a) shows the crossover to be quite nontrivial, being surprisingly abrupt for  $E_1^C$ .

*Parity effect.*—Whereas the ground state energies are not observable by themselves, the parity-dependent spectral gaps  $\Omega_0 = \mathcal{E}_2 - \mathcal{E}_0$  and  $\Omega_1 = \mathcal{E}_3 - \mathcal{E}_1$  are measurable in RBT's experiments by applying a magnetic field [8]. Figure 1(b) shows the canonical ( $\Omega_b^C$ ) and g.c. ( $\Omega_b^{GC}$ ) results for the spectral gaps. The main features of the g.c. predictions are as follows [8]: (i) The spectral gaps have a minimum, which (ii) is at a smaller  $d$  in the odd than the even case, and (iii)  $\Omega_1 < \Omega_0$  for small  $d$ , which was argued to constitute an observable parity effect. Remarkably, *the canonical calculation reproduces all of these qualitative features, including the parity effect*, differing from the g.c. case only in quantitative details: the minima are found at smaller  $d$ , and  $\Omega_0^{GC} < \Omega_0^C$  for large  $d$ . The latter is due to fluctuations, neglected in  $\mathcal{E}_b^{GC}$ , which are less effective in lowering  $\mathcal{E}_b^C$  the more levels are blocked, so that  $|\mathcal{E}_b^C - \mathcal{E}_b^{GC}|$  decreases with  $b$ .

*Wave functions.*—Next we analyze the variationally determined wave functions. Each  $|b\rangle$  can be characterized by a set of correlators:

$$C_j^2(d) = \langle c_{j+}^\dagger c_{j+} c_{j-}^\dagger c_{j-} \rangle - \langle c_{j+}^\dagger c_{j+} \rangle \langle c_{j-}^\dagger c_{j-} \rangle, \quad (4)$$

which measures the amplitude enhancement for finding a *pair* instead of two uncorrelated electrons in  $|j, \pm\rangle$ . For any blocked single-particle level and for all  $j$  of an uncorrelated state, one has  $C_j = 0$ . For the g.c. BCS case  $C_j = u_j v_j$  and the  $C_j$ 's have a characteristic peak of width  $\approx \tilde{\Delta}$  around  $\varepsilon_F$  [see Fig. 2(a)] implying that pairing correlations are “localized in energy space.” For the BCS regime  $d < \tilde{\Delta}$ , the canonical method produces  $C_j$ 's virtually identical to the g.c. case, *vividly illustrating why the g.c. BCS approximation is so successful: not performing the canonical projection hardly affects the parameters  $v_j$  if  $d \ll \tilde{\Delta}$ , but tremendously simplifies their calculation* (since the  $2n_0$  equations in (2) then decouple). However, in the f.d. regime  $d > d_b^C$ , the character of the wave function changes: weight is shifted into the tails far from  $\varepsilon_F$  at the expense of the vicinity of the Fermi energy. Thus *pairing correlations become delocalized in energy space* (as also found in [10]), so that referring to them as mere “fluctuations” is quite appropriate. Figure 2(b) quantifies this delocalization:  $C_j$  decreases as  $(A|\varepsilon_j - \varepsilon_F| + B)^{-1}$  far from the Fermi surface, with  $d$ -dependent coefficients  $A$  and  $B$ ; for the g.c.  $d = 0$  case,  $A = 2$  and  $B = 0$ ; with increasing  $d$ ,  $A$  decreases and  $B$  increases, implying smaller  $C_j$ 's close to  $\varepsilon_F$  but a *slower falloff far from  $\varepsilon_F$* . In the extreme case  $d \gg d_b^C$ , pair mixing is roughly equal for all interacting levels.

To quantify how the *total* amount of pairing correlations, summed over all states  $j$ , depends on  $d$ , Fig. 1(c) shows the *pairing parameter*  $\Delta_b(d) = \lambda d \sum_j C_j$

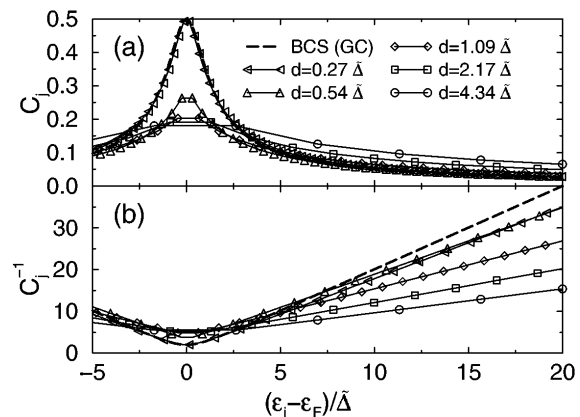


FIG. 2. The pairing amplitudes  $C_j$  of Eq. (4), for  $b = 0$ . (a) The dashed line shows the g.c. BCS result; pair correlations are localized within  $\tilde{\Delta}$  of  $\varepsilon_F$ . Lines with symbols show the canonical results for several  $d$ ; for  $d < d_0^C \approx 0.5\tilde{\Delta}$ , the wave functions are similar to the BCS ground state, while for  $d < d_0^C$  weight is shifted away from  $\varepsilon_F$  into the tails. (b) For all  $d$ ,  $C_j^{-1}$  shows linear behavior far from  $\varepsilon_F$ . For larger  $d$  the influence of levels far from  $\varepsilon_F$  increases.

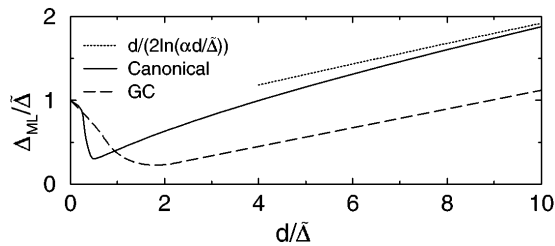


FIG. 3. The canonical (solid line), g.c. (dashed line), and perturbative (dotted line) results for the parity parameter  $\Delta_{ML}$  [9].

proposed by Ralph [8,16], calculated with the canonical ( $\Delta_b^C$ ) and grand-canonical ( $\Delta_b^{GC}$ ) approaches. By construction, both  $\Delta_b^{GC}$  and  $\Delta_b^C$  reduce to the bulk BCS order parameter  $\tilde{\Delta}$  as  $d \rightarrow 0$ , when  $C_j \rightarrow u_j v_j$ .  $\Delta_b^{GC}$  decreases with increasing  $d$  and drops to zero at the same critical value  $d_b^{GC}$  at which the energy  $E_b^{GC}$  vanishes [8], reflecting again the g.c. breakdown of superconductivity. In contrast,  $\Delta_b^C$  is nonmonotonic and never reaches zero; even the slopes of  $\Delta_e^C$  and  $\Delta_e^{GC}$  differ as  $d \rightarrow 0$  [5,6], illustrating that the  $1/N$  corrections neglected in the g.c. approach can significantly change the asymptotic  $d \rightarrow 0$  behavior (this evidently also occurs in Fig. 1b). Nevertheless,  $\Delta_b^C$  does show a clear remnant of the g.c. breakdown, by decreasing quite abruptly at the same  $d_b^C$  at which the plateau in  $E_b^C$  sets in. For the odd case this decrease is surprisingly abrupt, but is found to be smeared out for larger  $\lambda$ . We speculate that the abruptness is inversely related to the amount of fluctuations, which is reduced in the odd case by the blocking of the level at  $\varepsilon_F$ , but increased by larger  $\lambda$ .  $\Delta_b^C$  increases for large  $d$ , because of the factor  $\lambda d$  in its definition, combined with the fact that (unlike in the g.c. case) the  $C_j$  remain nonzero due to fluctuations.

Our quantitative analysis of the delocalization of pairing correlations is complimentary to but consistent with that of MFF [10]. Despite being limited to  $n_0 \leq 12$ , MFF also managed to partially probe the crossover regime from the f.d. side via an ingenious rescaling of parameters, increasing  $\lambda$  at fixed  $\omega_D$  and  $d$ , thus decreasing  $d/\tilde{\Delta}$ ; however, the total number of levels  $2\omega_d/d$  stays fixed in the process; thus this way of reducing the effective level spacing, apart from being (purposefully) unphysical, can yield only indirect and incomplete information about the crossover, since it captures only the influence of the levels closest to  $\varepsilon_F$ . Our method captures the crossover fully without any such rescalings.

*Matveev-Larkin's parity parameter.*—ML [9] have introduced a parity parameter, defined to be the difference between the ground state energy of an odd state and the mean energy of the neighboring even states with one electron added and one removed:  $\Delta_{ML} = \mathcal{E}_1 - \frac{1}{2}(\mathcal{E}_0^{\text{add}} + \mathcal{E}_0^{\text{rem}})$ . Figure 3 shows the canonical and g.c. results for  $\Delta_{ML}$ , and also the large- $d$  approximation given by ML,  $\Delta_{ML} = d/[2 \log(\alpha d/\tilde{\Delta})]$ , where the constant  $\alpha$  (needed

because ML's analysis holds only with logarithmic accuracy) was used as a fitting parameter (with  $\alpha = 1.35$ ). As for the spectral gaps, the canonical and g.c. results are qualitatively similar, though the latter, of course, misses the fluctuation-induced logarithmic corrections for  $d > d^C$ .

In summary, the crossover from the bulk to the f.d. regime can be captured in full using a fixed- $N$  projected BCS ansatz. With increasing  $d$ , the pairing correlations change from being strong and localized within  $\tilde{\Delta}$  of  $\varepsilon_F$ , to being mere weak, energetically delocalized fluctuations; this causes the condensation energy to change quite abruptly, at a characteristic spacing  $d^C \propto \tilde{\Delta}$ , from being *extensive* to *intensive* (modulo small corrections). Thus, the qualitative difference between superconductivity for  $d < d^C$ , and fluctuations for  $d > d^C$ , is that, for the former but not the latter, adding *more* particles gives a *different* condensation energy; for superconductivity, as Anderson put it, “more is different.”

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- [16]  $\bar{\Delta}_b \equiv \lambda d \sum_j \langle c_{j+}^\dagger c_{j-}^\dagger - c_{j-} c_{j+} \rangle \langle c_{j-} c_{j+} c_{j+}^\dagger c_{j-}^\dagger \rangle$ , an alternative pairing parameter proposed in [8], turns out to be identically equal to  $\Delta_b$  for the ansatz  $|b\rangle$ .