

## Exact study of the effect of level statistics in ultrasmall superconducting grains

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The reduced BCS model that is commonly used for ultrasmall superconducting grains has an exact solution worked out long ago by Richardson in the context of nuclear physics. We use it to check the quality of previous treatments of this model, and to investigate the effect of level statistics on pairing correlations. We find that the ground-state energies are on average somewhat lower for systems with nonuniform than uniform level spacings, but both have an equally smooth crossover from the bulk to the few-electron regime. In the latter, statistical fluctuations in ground-state energies strongly depend on the grain's electron number parity.

Recent experiments by Ralph, Black, and Tinkham, involving the observation of a spectroscopic gap indicative of pairing correlations in ultrasmall Al grains,<sup>1</sup> have inspired a number of theoretical<sup>2-11</sup> studies of how superconducting pairing correlations in such grains are affected by reducing the grains' size, or equivalently by increasing its mean level spacing  $d \propto \text{Vol}^{-1}$  until it exceeds the bulk gap  $\Delta$ . In the earliest of these, a grand-canonical (g.c.) BCS approach<sup>2-4</sup> was applied to a reduced BCS Hamiltonian for uniformly spaced, spin-degenerate levels; it suggested that pairing correlations, as measured by the condensation energy  $E^C$ , vanish abruptly once  $d$  exceeds a critical level spacing  $d^c$  that depends on the parity (0 or 1) of the number of electrons on the grain, being smaller for odd grains ( $d_1^c \approx 0.89\Delta$ ) than even grains ( $d_0^c \approx 3.6\Delta$ ). A series of more sophisticated canonical approaches (summarized below) confirmed the parity dependence of pairing correlations, but established<sup>6-11</sup> that the abrupt vanishing of pairing correlations at  $d^c$  is an artifact of g.c. treatments: pairing correlations do persist, in the form of so-called fluctuations, to arbitrarily large level spacings, and the crossover between the bulk superconducting (SC) regime ( $d \ll \Delta$ ) and the fluctuation-dominated (FD) regime ( $d \gg \Delta$ ) is completely smooth.<sup>10</sup> Nevertheless, these two regimes are qualitatively very different:<sup>9,10</sup> the condensation energy, e.g., is an extensive function of volume in the former and almost intensive in the latter, and pairing correlations are quite strongly localized around the Fermi energy  $\varepsilon_F$ , or more spread out in energy, respectively.

After the appearance of all these works, we became aware that the reduced BCS Hamiltonian on which they are based actually has an exact solution. It was published by Richardson in the context of nuclear physics (where it is known as the "picket-fence model"), in a series of papers between 1963 and 1977 (Refs. 12 and 13) which seem to have completely escaped the attention of the condensed-matter com-

munity. The beauty of this solution, besides its mathematical elegance,<sup>14</sup> is that it also works for the case of randomly spaced levels. It thus presents us with two rare opportunities that are the subject of this paper: (i) to compare the results of various previously used approximations against the benchmark set by the exact solution, in order to gauge their reliability for related problems for which no exact solutions exist; and very interestingly, (ii) to study the interplay of randomness and interactions in a nontrivial model *exactly*, by examining the effect of level statistics on the SC/FD crossover.

There is a previous study of the latter question by Smith and Ambegaokar (SA) using the g.c. mean-field BCS approach,<sup>5</sup> who concluded, interestingly, that randomness *enhances* pairing correlations: compared to the case of uniform spacings,<sup>2</sup> they found that a random spacing of levels (distributed according to the gaussian orthogonal ensemble) on average *lowers* the condensation energy  $E^C$  to more negative values and increases the critical level spacings at which  $E^C$  vanishes abruptly, but these still are parity dependent ( $\langle d_1^c \rangle = 1.8\Delta$ ,  $\langle d_0^c \rangle \approx 14\Delta$ ). However, the abrupt vanishing of  $E^C$  found by SA can be suspected to be an artifact of their g.c. mean-field treatment, as was the case in.<sup>2-4</sup> Indeed, our exact results for random levels show (1) that the SC/FD crossover is as smooth as for the case of uniformly spaced levels; this means, remarkably, that (2) even in the presence of randomness pairing correlations never vanish, no matter how large  $d/\Delta$  becomes; quite the opposite, (3) the randomness-induced lowering of  $E^C$  is strongest in the FD regime; in the latter, moreover, (4) the statistical fluctuations in  $E^C$  depend quite strongly on parity.

*Exact solution.* Ultrasmall superconducting grains are commonly described<sup>2-11</sup> by a reduced BCS model,

$$H = \sum_{j,\sigma=\pm} \varepsilon_{j\sigma} c_{j\sigma}^\dagger c_{j\sigma} - \lambda d \sum_{jj'} c_{j+}^\dagger c_{j-}^\dagger c_{j'-} c_{j'+}, \quad (1)$$

for a set of pairs of time-reversed states  $|j, \pm\rangle$  with energies  $\varepsilon_j$ , mean level spacing  $d$  and dimensionless coupling constant  $\lambda$ . Unbeknownst to the authors of Refs. 2–11, Richardson had long ago solved this model exactly, for an arbitrary set of levels  $\varepsilon_j$  (not necessarily all distinct): Since *singly occupied* levels do not participate in and remain “blocked”<sup>15</sup> to the pairscattering described by  $H$ , the labels of such levels are good quantum numbers. Let  $|n, B\rangle$  denote an eigenstate with  $N=2n+b$  electrons,  $b$  of which sit in a set  $B$  of singly occupied, blocked levels, thus contributing  $\mathcal{E}_B = \sum_{i \in B} \varepsilon_i$  to the total energy. The dynamics of the remaining  $n$  pairs is then governed by

$$H_B = \sum_{j \notin B} 2\varepsilon_j b_j^\dagger b_j - \lambda d \sum_{j, j' \notin B} b_j^\dagger b_{j'}, \quad (2)$$

where the pair operators  $b_j = c_{j-} c_{j+}$  satisfy the “hard-core boson” relations  $b_j^2 = 0$  and  $[b_j, b_{j'}^\dagger] = \delta_{jj'}(1 - 2b_j^\dagger b_j)$ , and the sums are over all *unblocked* levels. Richardson showed that the lowest-lying of the eigenstates  $|n, B\rangle$  has the (unnormalized) form (Ref. 14 gives a simple proof)

$$|n, B\rangle_G = \prod_{i \in B} c_{i\sigma}^\dagger \prod_{\nu=1}^n \left( \sum_{j \notin B} \frac{b_j^\dagger}{2\varepsilon_j - e_\nu} \right) |\text{Vac}\rangle, \quad (3)$$

where the  $n$  parameters  $e_\nu$  ( $\nu=1, \dots, n$ ) are that particular solution of the  $n$  coupled algebraic equations

$$\frac{1}{\lambda d} + \sum_{\mu=1(\neq \nu)}^n \frac{2}{e_\mu - e_\nu} = \sum_{j \notin B} \frac{1}{2\varepsilon_j - e_\nu} \quad (4)$$

that yields the lowest value for the “pair energy”  $\mathcal{E}(n) = \sum_{\nu=1}^n e_\nu$ . Moreover,  $|n, B\rangle_G$  has total energy  $\mathcal{E}(n) + \mathcal{E}_B$ . The lowest-lying of all eigenstates with  $n$  pairs and  $b$  blocked levels, say  $|n, b\rangle_G$  with energy  $\mathcal{E}_b^G(n)$ , is that  $|n, B\rangle_G$  for which the blocked levels in  $B$  are all as close as possible to  $\varepsilon_F$ , the Fermi energy of the uncorrelated  $N$ -electron Fermi sea  $|F_N\rangle$ .

In this paper we shall always take all the  $\varepsilon_j$  to be nondegenerate. The  $e_\nu$  then coincide at  $\lambda=0$  with the lowest  $n$  energies  $2\varepsilon_j$  ( $j=1, \dots, n$ ), and smoothly evolve toward lower values as  $\lambda$  is turned on. With increasing  $\lambda$ , the roots turn complex two at a time [becoming a complex conjugate pair, thus  $\mathcal{E}(n)$  remains real]. Denote roots destined to become conjugates by  $(e_{2a-1}, e_{2a})$  [with  $\lambda=0$  values  $(2\varepsilon_{j_{2a-1}}, 2\varepsilon_{j_{2a}})$ , say], with  $a=1, \dots, n/2$  for even  $n$ , with one further purely real root, say  $e_0$ , for odd  $n$ . Writing  $e_{2a-1} = \xi_a - i\eta_a$ ,  $e_{2a} = \xi_a + i\eta_a$ , they can be conveniently parametrized using the real variables  $x_a = \xi_a - \varepsilon_{2a-1} - \varepsilon_{2a}$  and  $y_a = -\eta_a^2 / [(\varepsilon_{2a} - \varepsilon_{2a-1})^2 - x_a^2]$ . When rewritten in terms of these, Eq. (4) becomes less singular [see Eq. (2.10) of Ref. 13 for details] and can easily be solved numerically by increasing  $\lambda$  from 0, using the set  $\mathcal{R} = \{(\varepsilon_{j_{2a-1}}, \varepsilon_{j_{2a}}), \varepsilon_{j_0}\}$  as “initial solution.”<sup>17</sup>

*Uniformly spaced levels.* Our first application of the exact solution is to check the quality of results previously obtained by various other methods. Most previous works<sup>2–4,6–10</sup> studied a half-filled band with fixed width  $2\omega_D$  of uniformly-spaced levels (i.e.,  $\varepsilon_j = jd$ ), containing  $N=2n+b$  electrons. Then the level spacing is  $d=2\omega_D/N$  and in the limit  $d \rightarrow 0$

the bulk gap is  $\Delta = \omega_D \sinh(1/\lambda)^{-1}$ . Following Ref. 9, we take  $\lambda=0.224$  throughout this paper. To study the SC/FD crossover, two types of quantities were typically calculated as functions of increasing  $d/\Delta$ , which mimics decreasing grain size: the even and odd ( $b=0,1$ ) condensation energies

$$E_b^C(n) = \mathcal{E}_b^G(n) - \langle F_N | H | F_N \rangle \quad (5)$$

and a parity parameter introduced by Matveev and Larkin<sup>6</sup> (ML) to characterize the even-odd ground-state energy difference,

$$\Delta^{\text{ML}}(n) = \mathcal{E}_1^G(n) - [\mathcal{E}_0^G(n) + \mathcal{E}_0^G(n+1)]/2. \quad (6)$$

Following the initial g.c. studies<sup>2–6</sup>, the canonical study of Mastellone, Falci, and Fazio,<sup>7</sup> (MFF) used Lanczos exact diagonalization (with  $n \leq 12$ ) and a scaling argument to probe the crossover regime. Berger and Halperin<sup>8</sup> (BH) showed that essentially the same results could be achieved with  $n \leq 6$  by first reducing the bandwidth and renormalizing  $\lambda$ , thus significantly reducing the calculational effort involved. To access larger systems and fully recover the bulk limit, fixed- $n$  projected variational BCS wave functions (PBCS) were used in Ref. 9 (for  $n \leq 600$ ); significant improvements over the latter results, in particular in the crossover regime, were subsequently achieved in Ref. 10 using the density-matrix renormalization group (DMRG) (with  $n \leq 400$ ). Finally, Dukelsky and Schuck<sup>11</sup> showed that a self-consistent random-phase approximation (RPA) approach, that in principle can be extended to finite temperatures, describes the FD regime rather well (though not as well as the DMRG).

To check the quality of the above methods, we<sup>16</sup> computed  $E_b^C(n)$  and  $\Delta^{\text{ML}}(n)$  using Richardson’s solution (Fig. 1). The exact results (a) quantitatively agree, for  $d \rightarrow 0$ , with the leading  $-\Delta^2/2d$  behavior for  $E_b^C(n)$  obtained in the g.c. BCS approach,<sup>2–4</sup> which in this sense is exact in the bulk limit, corrections being of order  $d^0$ ; (b) confirm that a completely smooth<sup>10</sup> crossover occurs around the scale  $d \approx \Delta$  at which the g.c. BCS approach breaks down; (c) show that the PBCS crossover<sup>9</sup> is qualitatively correct, but not quantitatively, being somewhat too abrupt; (d) are reproduced remarkably well by the approaches of MFF (Ref. 7) and BH;<sup>8</sup> (e) are fully reproduced by the DMRG of Ref. 10 with a relative error of  $< 10^{-4}$  for  $n \leq 400$ ; our figures do not show DMRG curves, since they are indistinguishable from the exact ones and are discussed in detail in Ref. 10.

The main conclusion we can draw from these comparisons is that the two approaches based on renormalization-group ideas work very well: the DMRG is essentially exact for this model, but the bandwidth rescaling method of BH also gives remarkably (though not quite as) good results with rather less effort. In contrast, the PBCS approach is rather unreliable in the crossover region.

*Randomly spaced levels.* The remainder of this paper addresses the question of how randomness of the levels  $\varepsilon_j$  affects pairing correlations. We studied half-filled bands of  $N=2n+b$  nonuniformly spaced but nondegenerate levels (for  $N \leq 260$ ), with  $b=0,1$ . The energy levels in small metallic grains with time reversal symmetry follow the Gaussian orthogonal ensemble distribution.<sup>18</sup> We generated sets of levels  $\varepsilon_i$  ( $i=1, \dots, N$ ) by diagonalizing  $2N \times 2N$  random matrices

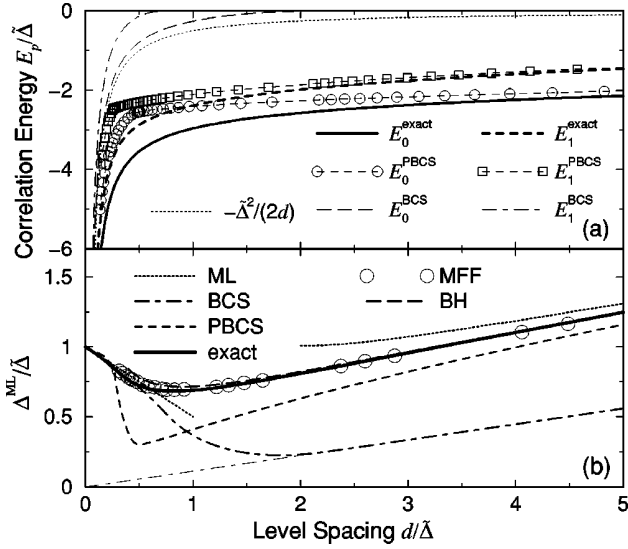


FIG. 1. (a) The even and odd ( $b=0,1$ ) condensation energies  $E_b^C$  of Eq. (5), calculated with BCS, PBCS, and exact wave functions, as functions of  $d/\Delta = 2 \sinh(1/\lambda)/(2n+b)$ , for  $\lambda = 0.224$ . For comparison the dotted line gives the ‘‘bulk’’ result  $E_0^{bulk} = -\Delta^2/(2d)$ . (b) Comparison of the parity parameters  $\Delta^{ML}$  (Ref. 6) of Eq. (6) obtained by various authors mentioned in the text. ML’s analytical result is  $\Delta(1-d/2\Delta)$  for  $d \ll \Delta$ , and  $d/2 \log(ad/\Delta)$  for  $d \gg \Delta$ , with  $a = 1.35$  adjusted to give asymptotic agreement with the exact result; for the grand-canonical BCS approach (dash-dotted line), the naive perturbative result  $\frac{1}{2}\lambda d$  is continued to the origin.

ces, taking  $N$  adjacent values from the central part of the eigenspectrum (to avoid boundary effects) and performing the rescaling<sup>5</sup>  $\varepsilon \rightarrow (1/2\pi)[4N \sin^{-1}(\varepsilon/\sqrt{4N}) + \varepsilon\sqrt{4N - \varepsilon^2}]$ , to ensure an average level spacing of one in units of  $d$ . In Fig. 2 we show four such sets of randomly generated levels for  $N=28$ , together with the equally spaced set.

For each such set of  $2n+b$  levels, we calculated the exact ground-state energy  $\mathcal{E}_b^G(n)$ , the condensation energy  $E_b^C(n)$ , and the spectroscopic gap<sup>4</sup>

$$E_b^S(n) = \mathcal{E}_{b+2}^G(n-1) - \mathcal{E}_b^G(n), \quad (7)$$

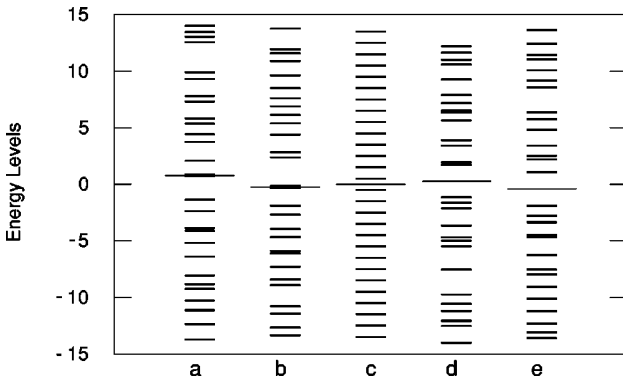


FIG. 2. Sets of energy levels with  $N=28$ . Set  $c$  has equally spaced levels, with spectroscopic gap [Eq. (7)]  $E_0^S/d = 1.54$ . Sets  $a, b$  (or  $c, d$ ) are randomly spaced; among all sets with  $N=28$  we studied, the ones shown have the smallest (largest) values for  $E_0^S/d$ , namely, 0.886, 0.891 (3.30, 3.37), due to the small (large) spacing between the two levels closest to  $\varepsilon_F$ , illustrating how random level fluctuations affect energy gaps.

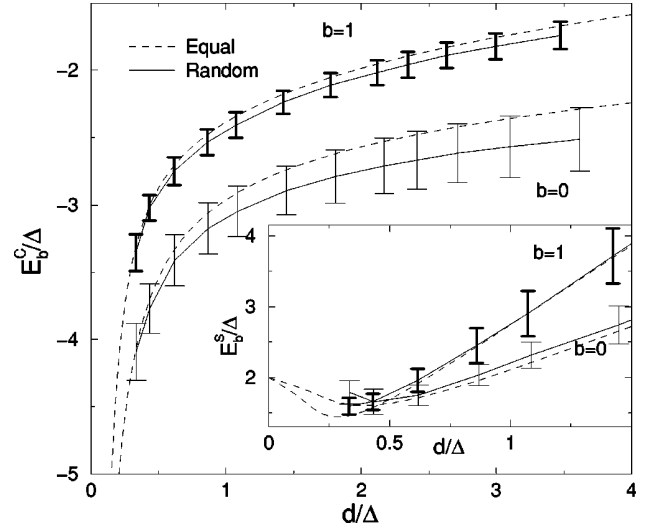


FIG. 3. Exact even and odd condensation energies  $E_b^C$  for equally spaced levels (dashed line), and the ensemble average  $\langle E_b^C \rangle$  for randomly spaced levels (solid line). The height of the fluctuation bars gives the variances  $\delta E_b^C$ . The inset shows the corresponding spectroscopic gaps  $E_b^S$  and variances  $\delta E_b^S$ .

which gives the energies needed to break a single pair in the (even or odd) ground state. Subsequently we calculated the ensemble average  $\langle E_b^C(n) \rangle$  and variance  $\delta E_b^C(n) = [\langle (E_b^C)^2 \rangle - \langle E_b^C \rangle^2]^{1/2}$  (and analogously  $\langle E_b^S \rangle$  and  $\delta E_b^S$ ) over many realizations of random matrices. The ensemble size was 1000 for  $24 \leq N \leq 40$ , and varied between 700 and 150 for  $40 \leq N \leq 260$ . Figure 3 presents our results for these ensemble averages (solid lines; fluctuation bars indicate variances) together with those for the uniformly spaced (u.s.) set discussed above (dashed lines). It shows a number of interesting features.

Firstly, the two main conclusions of SA (Ref. 5) are confirmed, namely (a) that pairing correlations are on average *stronger* for randomly than for uniformly spaced (u.s.) levels,  $\langle E_b^C \rangle < E_b^C(\text{u.s.})$ ; and (b) that the parity effect persists in the presence of randomness,  $\langle E_0^C \rangle < \langle E_1^C \rangle$ . In SA’s g.c. calculation these facts could be understood<sup>5</sup> from a condition, derived from the BCS gap equation, for having nonvanishing pairing correlations, namely  $2/\lambda < \sum_{j \in B} 1/|\bar{\varepsilon}_j - \bar{\mu}|$ . Here  $\bar{\varepsilon}_j$  and the g.c. chemical potential  $\bar{\mu}$  are in units of  $d$ , and the number of terms in the sum is of order  $2\omega_D/d$ . As  $d$  increases, this number decreases, until the inequality ceases to hold at a critical spacing  $d_b^c$ . Since statistical fluctuations to smaller values of  $|\bar{\varepsilon}_j - \bar{\mu}|$  carry more weight than those to larger values, fluctuations on average tend to increase  $d_b^c$ , which explains (a); moreover, since the blocking of levels close to  $\bar{\mu}$  reduces the number of terms in the sum, it reduces  $d_b^c$ , which explains (b).

Since the equation on which SA’s elegant argument is based breaks down in the FD regime, let us attempt another way of interpreting (a) and (b): pairing correlations involve a nonzero amplitude to find pair states with  $\varepsilon_j > \varepsilon_F$  doubly occupied and ones with  $\varepsilon_j < \varepsilon_F$  empty. Such correlations between states below and above  $\varepsilon_F$ , called ‘‘pair-mixing across  $\varepsilon_F$ ’’ in Ref. 2, gain interaction energy but cost some kinetic energy. The latter cost is the smaller, the closer the

states involved in pair-mixing across  $\varepsilon_F$  lie together (which is why the bulk limit  $d \rightarrow 0$  is so strongly correlated). Statistical fluctuations in level positions that yield more-closely or less-closely spaced levels around  $\varepsilon_F$  than for the uniform case, would thus cause a respectively lower or higher kinetic-energy cost for pairmixing across  $\varepsilon_F$ ; according to (a), the former on average outweighs the latter, just as SA had concluded in Ref. 5. Furthermore, in odd grains the blocked level at  $\varepsilon_F$  always causes the spacing between pair levels below and above  $\varepsilon_F$ , and hence the kinetic energy cost for pair mixing across  $\varepsilon_F$ , to be somewhat larger than in even grains, which explains (b).

Now, the ability of the exact solution to correctly treat the FD regime enables us to uncover several further facts that are beyond the reach of SA's g.c. mean-field approach: (c) The SC/FD crossover is as smooth for randomly as for uniformly spaced levels, confirming that the abrupt vanishing of pairing correlations at some critical level spacing found by SA is an artifact of their g.c. mean-field treatment, just as in Refs. 2 and 4. (d) Even in the presence of randomness, pairing correlations never vanish, no matter how large  $d/\Delta$ . Quite the opposite, (e) the randomness-induced lowering in condensation energy to more negative values,  $\langle E_b^C \rangle - E_b^C(\text{u.s.})$ , is *strongest* in the FD regime; this perhaps somewhat counter-intuitive result illustrates that the smaller the number of levels is that lie "close to" (i.e., within  $\Delta$  of)  $\varepsilon_F$ , the stronger is the effect of fluctuations in their positions on the kinetic-energy cost for pair mixing; conversely, this randomness-induced lowering of  $E_b^C$  decreases in the crossover regime and becomes negligible in the SC regime, in which very many levels lie within  $\Delta$  of  $\varepsilon_F$ . (f) The variances  $\delta E_b^C$  are

essentially  $d$  independent in the range  $24 \leq N \leq 260$ , implying that the *relative* statistical fluctuations of  $E_b^C$  should be negligible in the bulk limit, as expected.

Remarkably, we can also discern (g) three "parity-dependent fluctuation effects," in that the following three quantities are larger for even than for odd grains: (g1) the variances  $\delta E_b^C$  (with  $\delta E_0^C \approx 2 \delta E_1^C \approx \Delta/2$ ); and the randomness-induced changes in (g2) condensation energies  $|\langle E_b^C \rangle - E_b^C(\text{u.s.})|$  and (g3) spectroscopic gaps  $|\langle E_b^G \rangle - E_b^G(\text{u.s.})|$  (inset of Fig. 3). All three of these effects have the same origin as the more familiar parity effect (b), namely blocking: the more levels around  $\varepsilon_F$  are blocked, the larger the effective spacing between states involved in pair mixing across  $\varepsilon_F$ , and hence the smaller the sensitivity of the total energy to statistical fluctuations in level positions.

In conclusion, using Richardson's exact solution we have found that level randomness does not modify the smooth nature of the SC/FD crossover. It just enhances pairing correlations somewhat compared to those of uniformly spaced levels, having the strongest effect in the FD regime. In the latter we found that statistical fluctuations become strongly parity dependent.

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<sup>17</sup>For uniformly spaced levels, the  $\varepsilon_j$ 's are paired in ascending order,  $\mathcal{R} = \{(\varepsilon_1, \varepsilon_2), (\varepsilon_3, \varepsilon_4), \dots\}$ . For randomly spaced levels, two roots are the more likely to end up as conjugate pairs the closer they lie together at  $\lambda = 0$ . To construct  $\mathcal{R}$  for a given random spectrum of  $\varepsilon_j$ 's, we thus used the two neighboring levels with smallest spacing as  $(\varepsilon_{j(1)}, \varepsilon_{j(2)})$ , those with the next-smallest spacing as  $(\varepsilon_{j(3)}, \varepsilon_{j(4)})$ , and continued thus until no levels were left with spacings less than a certain cutoff  $\delta_\varepsilon$  ( $\leq d$ ). The remaining levels were paired in ascending order. We optimized  $\delta_\varepsilon$  (by trial and error) anew for each  $N$ . (An error in the choice of pairings in  $\mathcal{R}$  results in the failure of solutions with real  $x_a, y_a$  to exist beyond a certain  $\lambda$  value.)  
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