## Reduction of Electron Repulsion and Enhancement of  $T_c$  in Small Diffusive Superconducting Grains

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The superconducting properties of small metallic grains has been a topic of active research for half a century now. Early experiments demonstrated a remarkable rise in the critical temperature,  $T_c$ , with reducing grain size in a variety of materials. In two-dimensional diffusive superconductors,  $T_c$  is decreased due to enhanced Coulomb repulsion. We propose that in finite-size grains the diffusive enhancement of the Coulomb repulsion is weakened and leads ultimately to an increase in  $T_c$  in isolated, disordered two-dimensional grains. Our mechanism is superimposed on the possible enhancement in  $T_c$  due to the change in the density of states of finite-size systems.

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Introduction.—The superconducting properties of materials composed of small metallic grains has been a topic of enduring interest for more than five decades now, beginning with the pioneering theoretical work of Anderson in 1959 [\[1\].](#page-3-0) In the late 1960s, a series of experiments on thin films of granular Al, Sn, In, etc. [\[2,3\]](#page-3-1) found a remarkable enhancement in their transition temperatures as the grain size was reduced [\[4](#page-4-0)–6]. Later, improvements in electron tunneling methods enabled measurements on single grains [\[7\]](#page-4-1) and showed that the enhanced  $T_c$  in these grains was accompanied by an enhanced single particle gap compared to the bulk value. Recent experiments [\[8\]](#page-4-2) in dense grain arrays seem to be consistent with these older observations.

While initial explanations of this increase included proposals such as a surface enhancement of electron-phonon interactions [\[2\]](#page-3-1), later theories have tried to explain this in terms of various finite-size effects [\[9\]](#page-4-3), which become important with reducing grain size, as the single particle level spacing,  $\delta$ , increases. In relatively clean systems, this could lead to an enhancement in the DOS at the Fermi level, resulting in an increasing  $T_c$  with reducing size, until the grain becomes small enough such that  $\delta \sim \Delta$ , where  $\Delta$  is the superconducting gap. Below this minimum size, a coherent superconducting state can no longer be formed in a single grain, and the  $T_c$  disappears [\[1,10\]](#page-3-0). In dirty or irregularly shaped grains, on the other hand, the interplay of disorder, electron-electron repulsion, and finite-size effects brings nontrivial physics into play, as we explain in detail below.

In a conventional superconductor, the attractive interaction responsible for superconductivity is mediated by electron-phonon interactions. When an electron collides with a heavy ion, it distorts the ion from its equilibrium position. However, since the electron has an energy  $~\sim E_F$ , the Fermi energy, it escapes from the vicinity of the distortion in a time  $∼(h/E_F)$ , while it takes a much longer time ∼( $\hbar/\omega_D$ ), where  $\omega_D$  is the Debye energy, for the ion to relax. The distortion polarizes the metal, attracting other electrons to it. Crucially, because of the difference in time scales, a second electron attracted by the distortion experiences only a small repulsion from the initial one, which has escaped far away by that time, leading to an effective attraction between the two electrons. This reduction in the Coulomb repulsion between the two electrons can be formally expressed using various methods, including a renormalization group (RG) approach, leading to the wellknown Tolmachev-Anderson-Morel (TAM) logarithmic reduction [\[11\]](#page-4-4) in a clean system in the bulk.

In a diffusive system, the first electron escapes much more slowly since it collides frequently with impurities, and it may return to the original collision area. As a result, the reduction in the Coulomb repulsion is weaker than the clean case given by the TAM effect, causing a reduction in  $T_c$ . In two dimensions, this effect can be formulated in the RG language, leading to a modified RG equation below the scattering rate  $(1/\tau)$ , as shown by Finkelstein [\[12,13\]](#page-4-5).

In a finite-size system, the Thouless energy  $E_{\text{Th}} =$  $(\hbar D/L^2)$  defines another important energy scale. At energies much below  $E_{\text{Th}}$ , superconducting systems with dimensionless conductance  $g = (E_{\text{Th}}/\delta) \gg 1$  are described by Richardson's model [\[14\]](#page-4-6), with constant, energy-independent interaction matrix elements in the pairing channel. Physically, this expresses the fact that, at energies much below  $E_{\text{Th}}$ , the wave functions of all electrons are spread uniformly over the whole system, and thus the dynamical component of the electron-electron interaction is no longer present. In the RG language, this leads to the TAM logarithm again, resulting in a stronger reduction of the Coulomb interaction in this regime, similar to a clean system.

In a diffusive, finite-size grain, these energy scales form a hierarchy, given by  $E_F > 1/\tau > E_{Th}$ . Between  $E_F$  and  $1/\tau$ , the physics is identical to that of a bulk clean system since the electrons are unaware of the disorder and the finite size. Hence, the RG is determined by the TAM equation. Between  $1/\tau$  and  $E_{\text{Th}}$ , the electrons are affected by disorder but not the finite size and thus follow the Finkelstein equation. However, below  $E_{\text{Th}}$ , the finite-size effect dominates, and the RG reverts to the TAM equation due to the arguments provided above.

As the grain size L is reduced,  $E_{\text{Th}}$  is increased, diminishing the regime where the Finkelstein effect is relevant, while simultaneously extending the regime where the TAM equation holds. As a result, in a smaller grain, the Coulomb repulsion is reduced more strongly since the Finkelstein regime is smaller, and this should lead to a larger mean field  $T_c$ .

These conclusions are confirmed by our calculations. We consider isolated grains with size  $L \gg t$ , where t is the thickness of the grains, and study the mean field  $T_c$  as a function of  $E_{\text{Th}}$  by solving the appropriate RG equations in the different regimes. Using a specific model for the bare interactions based on the physical arguments given above, we show that the mean field  $T_c$  can be increased all the way from the disordered bulk limit,  $T_c^b$ , to the clean limit,  $T_{c0}$ , by simply reducing the size of the grain such that  $T_c^b < E_c < 1/\tau$ , where  $E_c = 4\pi^2 E_{\text{Th}}$ . We observe an increase of up to 20% in  $T_c$  when  $g \sim O(10)$ , but at the limit of the validity of the theory, when  $g \sim O(1)$ , T<sub>c</sub> increases by up to 60%.

RG in clean systems.—Superconductivity is driven by a diverging interaction in the pairing channel or Cooper channel. In a clean system, the physics is contained in the repeated scattering of electrons with opposite momenta, Matsubara frequencies, and spin,  $|\vec{k}, \epsilon_m \uparrow \rangle$  and  $|-\vec{k}, -\epsilon_m \downarrow \rangle$ , by the interaction [\[15\]](#page-4-7). The screened Coulomb interaction  $V_{\text{scr}}$  is usually assumed to be local and instantaneous, and hence the matrix elements for pair scattering between states  $|\vec{k}, \epsilon_m \uparrow \rangle$  and  $|-\vec{k}, -\epsilon_m \downarrow \rangle$  and  $|\vec{k'}, \epsilon_n \uparrow \rangle$  and  $|-\vec{k'}, -\epsilon_n \downarrow \rangle$  is given by a constant  $\Gamma^0_{mn} = \nu_0 V_{\rm scr} \approx 1$ , where  $\nu_0$  is the density of states at the Fermi energy.

The effective phonon-mediated attractive interaction below  $\omega_D$  is retarded, and hence frequency dependent, in general, but it is assumed to be a constant for simplicity. Denoting its value by  $\lambda_a$ , the bare matrix elements in the clean system are given by

$$
\Gamma_{mn}^0 = 1, \quad E_F > \max(\epsilon_m, \epsilon_n) > \omega_D
$$

$$
= (1 - \lambda_a), \quad \epsilon_m, \epsilon_n < \omega_D.
$$
 (1)

<span id="page-1-3"></span>The full effective interaction  $\Gamma_{mn}$  can be found by solving the relevant Bethe-Salpeter equation (see the discussion on the disordered case below), or by progressively integrating out thin regions of energy in succession. With  $\epsilon_m, \epsilon_n \to \omega$  and  $\Gamma_{mn} \equiv \Gamma(\omega)$ , both methods lead to the standard TAM RG equation [\[11\],](#page-4-4) given by

$$
d\Gamma(\omega)/dl_{\omega} = -\Gamma^2(\omega). \tag{2}
$$

<span id="page-1-0"></span>Here,  $\omega$  is the running energy scale and  $l_{\omega} = \ln(E_F/\omega)$ . Notice that the bare matrix elements do not appear in the equation directly, but only through the boundary condition  $\Gamma(E_F) = \Gamma_0(E_F) \approx 1$ . This is easily integrated to give the TAM logarithm reduction [\[11\]](#page-4-4)

$$
\Gamma(\omega) = \Gamma(E_F) / [1 + \Gamma(E_F) \ln(E_F/\omega)].
$$
 (3)

Now the RG proceeds in two steps. First, from  $E_F$  to  $\omega_D$ , the RG reduces the effective interaction strongly, according to Eq. [\(3\).](#page-1-0) If the attractive interaction due to the phonons,  $\lambda_a$ , is stronger than the renormalized repulsive interaction  $\Gamma(\omega_D)$ , then the total interaction is negative and further renormalization increases until it diverges at  $\omega = T_c \sim$  $\omega_D \exp[-1/|\Gamma(\omega_D) - \lambda_a|]$ . In some places in the literature [\[16\]](#page-4-8),  $\Gamma(\omega_D)$  is denoted by  $\mu^*$ .

Disordered.—In disordered systems, one should take into account corrections to the  $\Gamma_{mn}$  due to disorder. At weak potential disorder,  $1/\tau \ll \omega_D$ , it is well known that the superconducting  $T_c$  is virtually unchanged [\[1\]](#page-3-0). Diagrammatically, this corresponds to incorporating the effects of disorder and interactions to  $\Gamma_{mn}$  separately [\[13\]](#page-4-9), i.e., where the electron-electron interactions and disorder corrections are factorizable.

However, there is a class of corrections that couples different sections of the matrix elements with different indices  $m$  and  $n$ . These provide a nontrivial frequency dependence to the resulting matrix elements, and the disorder and interaction corrections are no longer factorizable. While these corrections are minor in three dimensions, they become important in lower dimensions. In two dimensions, the disorder corrected bare Coulomb matrix elements  $\Gamma_{mn}$  can be explicitly calculated by diagrammatic methods and are given by [\[13,17\]](#page-4-9)

<span id="page-1-1"></span>
$$
\Gamma_{mn}^0 = 1 + ut \ln[1/(\epsilon_m + \epsilon_n)\tau], \qquad \epsilon_n, \epsilon_m < 1/\tau. \tag{4}
$$

Here,  $\epsilon_m$  and  $\epsilon_n$  are fermionic Matsubara frequencies,  $u \sim$ 0.5 in two dimensions and  $t = (1/2\pi^2)(e^2/\hbar)R_{\Box}$  $1/(2\pi^2 g)$ , where  $R_{\Box}$  is the sheet resistance in two dimensions. The first term is the contribution from the clean bare matrix element, while the frequency dependent second term encodes the contribution from disorder corrections.

<span id="page-1-2"></span>This form can also be derived by calculating the pairing channel matrix elements between exact disorder eigenstates  $|m \uparrow \rangle$ ,  $|m \downarrow \rangle$  and  $|n \uparrow \rangle$ ,  $|n \downarrow \rangle$ . Using semiclassical arguments [\[18\]](#page-4-10), one can show that

$$
\nu_0 V_{mn} = \sum_{q, Dq^2 < 1/\tau} |\langle m| e^{iqr} |n \rangle|^2
$$
\n
$$
= (\delta/\pi) \sum_{q, Dq^2 < 1/\tau} Dq^2 / (D^2 q^4 + \omega_{mn}^2). \tag{5}
$$

Here,  $\omega_{mn}$  is the energy difference between the states m and  $n$ . In the continuum limit in two dimensions, the <span id="page-2-0"></span>resultant integral yields the logarithm of Eq. [\(4\).](#page-1-1) The full matrix element  $\Gamma_{mn}$  is given by the following Bethe-Salpeter equation [\[17\]](#page-4-11):

$$
\Gamma_{mn} = \Gamma^0_{mn} - 2\pi T \sum_r \Gamma^0_{mr} \frac{1}{\varepsilon_r} \Gamma_{rn}.
$$
 (6)

<span id="page-2-1"></span>The logarithmic behavior of  $\Gamma^0_{mn}$  enables a RG treatment of the system using the maximum section approach [\[19\]](#page-4-12) with the approximation  $\ln[(\epsilon_m + \epsilon_n)\tau] \approx \ln(\max[\epsilon_m, \epsilon_n]\tau)$ . One then gets a modified RG equation for  $\Gamma(\omega)$  given by [\[13,17,20\]](#page-4-9)

$$
d\Gamma(\omega)/dl_{\omega} = ut - \Gamma^2(\omega). \tag{7}
$$

The first term is the nontrivial contribution due to disorder and slows down the renormalization of the scattering amplitude. In a thermodynamic two-dimensional system, this leads to a suppression of the  $T_c$  with increasing disorder.

*Finite size*.—In finite-size systems,  $E_{\text{Th}}$  provides another important energy scale in addition to  $E_F$ ,  $1/\tau$ ,  $\omega_D$ , and  $T_c$ , as explained earlier. It is well known that the statistical properties of the energy eigenstates of such systems for energy scales  $\omega \ll E_{\text{Th}}$  and  $g \gg 1$  are described by random matrix theory [\[22\].](#page-4-13) Under these conditions, the system is described by the so-called universal Hamiltonian [\[23\],](#page-4-14) which is determined by three constant coefficients coupling to the total density, spin, and pairing operators, respectively. In situations where only the pairing channel is relevant, this reduces to the well-known Richardson model [\[14\]:](#page-4-6)

$$
\mathcal{H}_{\text{Rich}} = \sum_{m,\sigma} \epsilon_m c_{m\sigma}^{\dagger} c_{m\sigma} + \lambda \delta \sum_{m,n} c_{m\uparrow}^{\dagger} c_{m\downarrow}^{\dagger} c_{n\downarrow} c_{n\uparrow}.
$$
 (8)

Here, m, n denote the exact eigenstates of the system and λ encodes the strength of the electron-electron interactions. Thus, the matrix elements become independent of the states they couple, similar to the clean case, and this leads to the TAM RG equations. To come up with a specific model for the bare matrix elements, we make the crude assumption that we can neglect the frequency dependence of the elements below  $\omega < Dq_{\min}^2 = E_c$  in Eq. [\(5\).](#page-1-2) Hence, for  $\omega < E_c$ , the bare Coulomb matrix elements assume the constant value  $\lambda \sim 1 + ut \ln[1/(E_c \tau)]$ .  $E_c$  acts as an effective cutoff scale for the bare matrix elements and plays a fundamental role in the scaling properties of the system.

Hence, in superconducting grains with a relatively large  $E_{\text{Th}}$  and  $L \gg t$ , there are three distinct regimes:

(1)  $E_F > \omega > 1/\tau$ . The system is in the ballistic limit, the bare Coulomb matrix elements are given by  $\Gamma_{mn}^0 = \lambda_0 = 1$ , and the full matrix element  $\Gamma$  follows the TAM RG equation, Eq. [\(4\)](#page-1-1).

(2)  $1/\tau > \omega > E_c$ . In this regime, the Coulomb repulsion is affected by disorder,  $\Gamma_{mn}^0$  is given by Eq. [\(4\),](#page-1-1) and the RG is given by the Finkelstein equation, Eq. [\(6\)](#page-2-0).

(3)  $E_c > \omega > T_c$ . In this regime, the matrix elements are effectively constant and the system can be described by the Richardson model. Using our crude model described earlier,  $\Gamma_{mn}^0 \sim 1 + ut \ln[1/(E_c \tau)]$ , and the system again follows the TAM RG equation.

Of course, for all cases where  $(\epsilon_m, \epsilon_n) < \omega_D$ , the total bare matrix element also includes the attractive interaction,  $(-\lambda_a)$ .

These considerations lead to a remarkable conclusion: In the regime  $1/\tau > E_c > T_c$ , increasing  $E_c$  by reducing the size of the system diminishes regime 2 and simultaneously extends regime 3, resulting in a *faster* renormalization of the effective interaction. This will lead to an increase in the mean field  $T_c$  of the system, until  $E_c = 1/\tau$ , where  $T_c$  will be equal to the clean limit value of the material, since the RG would then be determined by the TAM equations throughout the whole energy range. Thus, within mean field theory, one can increase  $T_c$  all the way from the bulk disordered value,  $T_c^b$  [given by the solution of Eq. [\(7\)\]](#page-2-1), to the clean value  $T_c^0$ . As mentioned before, this picture ceases to be valid when  $\delta \sim \Delta \sim O(T_c)$ , where the superconducting state ceases to exist.

Numerical results.—We consider isolated grains of materials with transverse dimensions L and thickness  $t \ll L$ , so that they are effectively two dimensional. For direct comparison with real materials, we choose our energy unit such that important parameters like  $E_F$  assume values similar to those in real materials expressed in kelvins (K). We choose reasonable values:  $E_F = 30000 \text{ K}$ ,  $1/\tau = 3000 \text{ K} = 0.1E_F$ , and  $\omega_D = 300 \text{ K} = 0.01E_F$ . Using typical values for density of states  $\nu_0$  and Fermi velocity  $v_F$  for free electrons, we find a mean free path  $l_e \approx 2.5$  nm. Note that, for a system with  $t = l_e$ ,  $g = (E_{\text{Th}}/\delta) = \hbar D v_0 t \approx 9$ . The maximum value of  $E_c = (1/\tau)$  implies that the corresponding minimum length  $L = \sqrt{4\pi^2 \hbar D \tau} \approx 9.1$  nm.

First, we analyze the behavior of the bare matrix elements to gain insight into the RG process. Figure [1](#page-3-2) shows the bare Coulomb matrix elements, as is explained in the figure caption in detail. The plots demonstrate how these elements, which increase logarithmically for  $\omega > E_c$ , essentially saturate below this value, validating our crude assumptions in the previous section. We have omitted the attractive interaction below  $\omega_D$  in the plots for clarity.

Figure [2](#page-3-3) shows our primary result: the normalized transition temperature  $T_c$  with an increasing  $E_c$  for two cases, with  $T_{c0} = 7$  K and 1 K, respectively, crudely corresponding to materials with a moderately large  $T_c$ such as Mo-Ge [\[24\]](#page-4-15) and small  $T_c$  such Al. We choose the same set of parameters for both except for the attractive interaction parameter  $\lambda_a$ , which is adjusted to yield the respective values of  $T_c$ . We find that at large g (∼10), the system with the larger  $T_c$  shows an increase of 12%, while the one with the smaller  $T_c$  shows a much larger increase of 20%. Hence, grains with a small  $T_c$  show a much larger fractional increase with reducing size at large  $g$ , which is a

<span id="page-3-2"></span>

FIG. 1. Bare Coulomb matrix elements  $\Gamma^0$  vs frequency  $\omega$  for different values of  $E_c = 4\pi^2 E_{\text{Th}}$  (increasing from the topmost curve downwards), where  $E_{\text{Th}}$  is the Thouless energy, at dimensionless conductance  $g = 10$  and scattering rate  $1/\tau = 0.1E_F$ , where  $E_F$  is the Fermi energy. All energy scales are expressed in units of 1 K, which is defined in detail in the text. The matrix elements show logarithmic behavior for  $\omega \gtrsim E_c$ , consistent with Eq. [\(4\)](#page-1-1), but saturate to a constant below this energy scale as it enters the Richardson regime, as explained in the text. The corresponding RG equations in these regimes are given by Eqs. [\(4\)](#page-1-1) and [\(2\),](#page-1-3) respectively, as explained in detail in the text.

correlation borne out by experiments. Furthermore, by pushing the theory close to its limit of validity  $q \sim O(1)$ , we get an enhancement close to 60% in both cases. We reiterate that this remarkable conclusion follows simply from examining the RG flow of the system with various

<span id="page-3-3"></span>

FIG. 2. Normalized  $T_c$  with increasing  $E_c = 4\pi^2 E_{\text{Th}}$ , where  $E_{\text{Th}}$  is the Thouless energy, for two different systems with clean limit transition temperatures  $T_{c0} = 7$  K (left column) and  $T_{c0} =$ 1 K (right column) at different values of dimensionless conductance  $q$  (increasing from the topmost curve downwards). All energy scales are expressed in units of 1 K, which is defined in detail in the text. The x-axis cutoff is determined by  $\delta \sim T_c$ , beyond which superconductivity should not survive [\[1,10\]](#page-3-0). At large  $g \sim O(10)$ , the small  $T_c$  system shows a much larger fractional increase (20%) than the one with a large  $T_c$  (12%). Close to the limit of the validity of the theory, at  $g \sim O(1)$ , the enhancement can increase up to 60%.

values of  $E_c$ , with no reference whatsoever to specific details of the material parameters and its geometry.

Experimental verification and discussion.—Our theory concerns isolated grains, studied in some experiments [\[25\]](#page-4-16), whereas other experimental samples consist of an array of such grains coupled by an effective Josephson coupling. This provides a new energy scale in these systems whose collective properties, including transport, may be very different from those of individual grains. Hence, to verify our predictions experimentally, one must focus on isolated or weakly coupled grains, and measurements sensitive to the single particle indicators of the superconducting state of individual grains, such as the local gap. Examples of the above are the specific heat capacity, which shows a peak at the superconducting transition, and scanning tunneling measurements, which can measure the local density of states, and thus the local gap, directly. Hence, we propose that our predictions should be verifiable from measurements of specific heat capacity and tunneling spectra to track the transition in individual grains.

As discussed earlier, we have neglected various other finite-size effects discussed in the literature that may lead to an enhancement of the  $T_c$  in relatively clean grains. Since our mechanism is completely different from these, our effect will be superposed on all these in real disordered grains. This seems to be relevant in granular Al films, for example, where the  $T_c$  can substantially exceed  $T_{c0}$  [\[8\],](#page-4-2) but here our mechanism may contribute a part of the total increase. Furthermore, we have neglected the progressive broadening of the superconducting transition with an increasing  $\delta/T_c$  (leading to its eventual disappearance in the  $\delta/T_c \rightarrow 1$  limit [\[1,10\]\)](#page-3-0), analyzed in detail in Ref. [\[26\]](#page-4-17). These results seem to indicate that the broadening is not accompanied by an appreciable shift in the transition in small grains. We have also not discussed the effects of fluctuations in the grain sizes in experiments on distributions of grains [\[27\]](#page-4-18).

In conclusion, by considering the RG equations in different energy regimes, we have demonstrated a universal mechanism for increasing the superconducting  $T_c$  in isolated disordered grains with reducing size, from the bulk value  $T_c^b$  to the clean limit  $T_{c0}$  in the regime  $T_c < E_c < 1/\tau$ . This prediction can be tested experimentally by measuring properties sensitive to the local single particle gap such as the specific heat capacity.

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