Evolution from BCS superconductivity to Bose condensation: Role of the parameter $k_F\xi$

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We argue that the natural variable to establish the crossover from Cooper-pair-based superconductivity to Bose-Einstein condensation of bound-electron pairs is the product $k_F\xi$ of Fermi electron wave vector times the coherence length for two-electron correlation, and that Cooper-pair-based superconductivity is stable against bosonization down to $k_F\xi \simeq 2\pi$. We also propose that the experimental plot by Uemura *et al.*, relating the superconducting temperature T_c to the Fermi temperature T_F for a variety of "exotic" as well as conventional superconductors, can be rationalized by correlating the ratio T_c/T_F to $k_F\xi$. It appears in this way that the high-T_c superconductors lie in the plot by Uemura *et al.* near the "instability" line $k_F\xi = 2\pi$.

Uemura *et al.*[1] have recently proposed to distinguish a class of "exotic" superconductors (including cuprate, bismuthate, organic, Chevrel-phase, heavy fermions, and fullerene systems) from the more conventional superconductors (like Nb) by the value of the ratio T_c/T_F between the experimental superconducting temperature and the (effective) Fermi temperature, which turns out to be about one hundred times larger for the "exotic" than for the conventional superconductors. Uemura *et al.* have also suggested that this substantial difference might be an indication that the "exotic" superconductors are in some sense intermediate between conventional BCS superconductors and Bose-condensed systems.

In order to establish a possible connection between the physical consequences of this suggestion and the experimental relation between T_c and T_F, one should first try to figure out the appropriate variable to determine the crossover between Cooper-pair-based (BCS) superconductivity and Bose-Einstein (BE) condensation. One could then attempt to use that variable as a sort of "normal" coordinate in the plot constructed by Uemura *et al.* (hereafter referred to simply as the Uemura plot), by figuring out a phenomenological relation (independent of the underlying superconducting mechanism) between that variable and the ratio T_c/T_F, thus encompassing the difference between the "exotic" and conventional superconductors. Completion of this program might further turn out to be useful both experimentally (by suggesting possibly in which direction one should move in the Uemura plot to improve T_c) and theoretically (by giving hints on the underlying dynamics related to high-T_c superconductivity). In this paper we shall propose how to meet this program by a rather general argument on the stability of the Cooper-pair-based superconductivity.

Evolution from weak- to strong-coupling superconductivity has been addressed a few years ago by Nozières and Schmitt-Rink[2] (hereafter referred to as NSR) following the pioneering work by Legget.[3] Central to this work is the well-known argument[4] that the BCS wave function has built in the Bose-Einstein condensation as a limiting case, since it reduces to the BE condensate wave function when the (average) occupation numbers $\langle n_{\mathbf{k},\sigma}\rangle$ can be neglected with respect to unity for *all* wave vectors $\mathbf{k}$ (and for both spin projections σ). NSR follow the evolution from Cooper-pair-based superconductivity to BE condensation through the increase of the coupling strength associated to an effective fermionic attractive potential, and conclude that the evolution is "smooth." Although this result is appealing from a theoretical point of view, it does not allow for a direct comparison with the Uemura plot since the coupling strength of the effective fermionic attraction is not a quantity that could be realistically inferred from experiments. Besides, associating an effective coupling strength to a given class of superconductors would unavoidably require one to face at the outset the problem of the mechanism responsible for superconductivity,[5] which need not be actually necessary to unravel the kind of message conveyed by the Uemura plot.

We need thus to figure out a more significant variable than the coupling strength to follow the evolution from BCS superconductivity to BE condensation. The new variable should be selected according to the following criteria: (i) the evolution from BCS to BE should turn out to be as much as possible *universal*, i.e., independent of the details of the interaction potential and of the single-particle density of states; (ii) when using the new variable to rationalize the Uemura plot, it should be possible to subject that variable to an *independent* experimental check.[6] We shall illustrate in the following why we propose to identify the product $k_F\xi$ as the desired variable.

Following NSR, we introduce the model fermionic

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Hamiltonian

$$H=\sum_{\mathbf{k},\sigma}\epsilon_{\mathbf{k}}c^{\dagger}_{\mathbf{k},\sigma}c_{\mathbf{k},\sigma}+\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}}V_{\mathbf{k},\mathbf{k}'}c^{\dagger}_{\mathbf{k}+\mathbf{q}/2,\uparrow}c^{\dagger}_{-\mathbf{k}+\mathbf{q}/2,\downarrow}\times c_{-\mathbf{k}'+\mathbf{q}/2,\downarrow}c_{\mathbf{k}'+\mathbf{q}/2,\uparrow}\ , \tag{1}$$

where $c_{\mathbf{k},\sigma}$ is the destruction operator for fermions with wave vector $\mathbf{k}$ and spin σ, $\epsilon_{\mathbf{k}}$ is a single-particle (or quasiparticle) dispersion relation, and $V_{\mathbf{k},\mathbf{k}'}$ is an "effective" fermionic attraction. In its simplest version, $\epsilon_{\mathbf{k}}=\mathbf{k}^2/2m^*-\mu$ where m^* is an effective (quasi)particle mass and μ is the chemical potential.[7] Although the use of the Hamiltonian (1) has obvious shortcomings, by resting on a continuum model (where no effect of the lattice structure is included) and by disregarding dynamical effects, we believe that it is sufficient for our purposes.

The variational procedure with the usual BCS trial wave function

$$|\Phi\rangle=\prod_{\mathbf{k}}(u_{\mathbf{k}}+v_{\mathbf{k}}c^{\dagger}_{\mathbf{k},\uparrow}c^{\dagger}_{-\mathbf{k},\downarrow})|0\rangle \tag{2}$$

leads to the two familiar *coupled* equations

$$2\epsilon_{\mathbf{k}}\phi_{\mathbf{k}}+(1-2v_{\mathbf{k}}^2)\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\phi_{\mathbf{k}'}=0\ , \tag{3}$$

$$n=\frac{1}{\Omega}\sum_{\mathbf{k}}\left[1-\frac{\epsilon_{\mathbf{k}}}{E_{\mathbf{k}}}\right]\ , \tag{4}$$

where n is the particle density, Ω the quantization volume, and $\phi_{\mathbf{k}}=2u_{\mathbf{k}}v_{\mathbf{k}}=\Delta_{\mathbf{k}}/E_{\mathbf{k}}$ with $\Delta_{\mathbf{k}}=-\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}u_{\mathbf{k}'}v_{\mathbf{k}'}$ and $E_{\mathbf{k}}=\sqrt{\epsilon_{\mathbf{k}}^2+\Delta_{\mathbf{k}}^2}$.[8] Note that, provided $v_{\mathbf{k}}^2\ll 1$ for *all* $\mathbf{k}$, Eq. (3) reduces to the Schrödinger equation for the relative motion of two particles with equal mass m^* interacting via $V_{\mathbf{k},\mathbf{k}'}$ and with eigenvalue 2μ. In this limit bosonization of bound-electron pairs is fully achieved.

Solution of Eqs. (3) and (4) gets considerably simplified by considering a separable potential. NSR choose

$$V_{\mathbf{k},\mathbf{k}'}=Vw_{\mathbf{k}}w_{\mathbf{k}'},\quad w_{\mathbf{k}}=\frac{1}{\sqrt{1+(\mathbf{k}/k_0)^2}}\ , \tag{5}$$

where the strength $V(<0)$ and the characteristic wave vector k_0 are the parameters of the interaction. With this potential, $\Delta_{\mathbf{k}}=\Delta_0 w_{\mathbf{k}}$ and the associated two-body eigenvalue problem has (in three dimensions) the only eigenvalue $2\mu=-\epsilon_0=-(k_0^2/m^*)(G-1)^2$ for $G>1$, where $G=-V\Omega m^*k_0/4\pi>0$ is the dimensionless coupling constant, while the bound-state radius has the asymptotic behavior $r_0\sim k_0^{-1}G^{-1/2}$ for $G\to\infty$.[9] In agreement with the results by NSR, we find that the solutions (Δ_0,μ) of Eqs. (3) and (4) evolve smoothly as functions of G *for given* k_0, although (Δ_0,μ) depend strongly on k_0 for given G.[10] No connection with the Uemura plot is evidently possible at this level.

A suggestion to figure out a more significant variable than the coupling strength comes from the observation that "exotic" superconductors of the Uemura plot have a considerable shorter *coherence length* ξ ($\sim$20–50 Å) than the conventional superconductors (for which $\xi\sim 10^3$–10^4 Å). Theoretically, ξ can be obtained from the pair-correlation function with opposite spins

$$g(\mathbf{r})=\frac{1}{n^2}|\langle\Phi|\Psi^{\dagger}_{\uparrow}(\mathbf{r})\Psi^{\dagger}_{\downarrow}(0)|\Phi\rangle|^2\ , \tag{6}$$

where $\Psi_\sigma(\mathbf{r})$ is the fermion field operator, by identifying

$$\xi^2=\frac{\int d\mathbf{r}\,g(\mathbf{r})\mathbf{r}^2}{\int d\mathbf{r}\,g(\mathbf{r})}=\frac{\sum_{\mathbf{k}}|\nabla_{\mathbf{k}}\phi_{\mathbf{k}}|^2}{\sum_{\mathbf{k}}|\phi_{\mathbf{k}}|^2}\ . \tag{7}$$

Our definition (7) correctly reduces to the Pippard coherence length $\xi_0=(d\epsilon_{\mathbf{k}}/dk)_{k_F}/\pi\Delta_{k_F}$ in the weak-coupling limit (actually, $\xi\to\xi_0\pi/2\sqrt{2}=1.11\xi_0$ when $G\ll 1$) and to the bound-state radius r_0 in the strong-coupling limit. It turns out that the behavior of ξ versus G is also "smooth," although it strongly depends on the parameter k_0 of the interaction. A reasonable attempt to eliminate the coupling constant G from further considerations is then to replace it by a dimensionless parameter containing ξ. Since k_F^{-1} is the only other independent physical length scale in the problem, we replace the original pair of variables (G,k_0) by the alternative pair $(k_F\xi,k_0)$ and study the crossover from BCS superconductivity to BE condensation as a function of $k_F\xi$ for given k_0.[11]

In Fig. 1 we report the chemical potential μ versus $k_F\xi$ for a wide range of values of the reduced density n/k_0^3 $(=10^\alpha$ with $\alpha=-5,-4,\ldots,+4)$. Positive values of μ have been normalized by the Fermi energy ϵ_F $(=k_F^2/2m^*$, by our definition), while negative values of μ have been normalized by half of the eigenvalue ϵ_0 of the

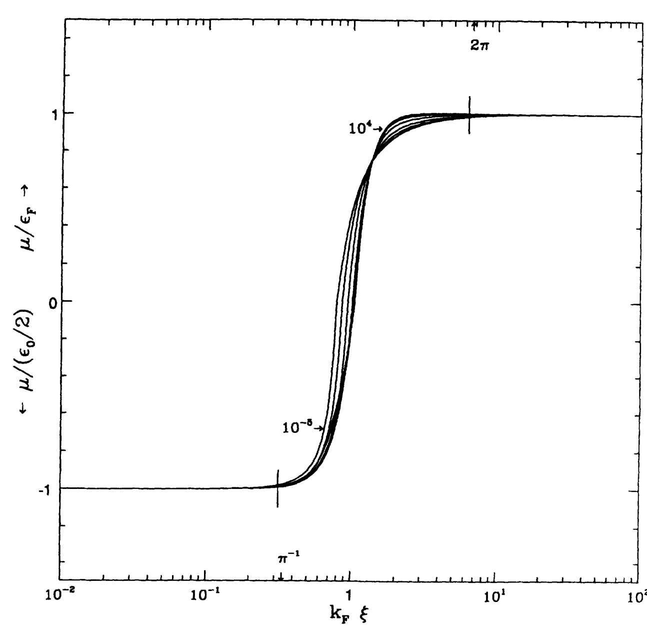

FIG. 1. Chemical potential μ versus $k_F\xi$ (at zero temperature). The normalization of μ and the meaning of the different curves are explained in the text. The two limiting curves corresponding to the values 10^{-5} and 10^4 of the reduced density n/k_0^3 are indicated.

associated two-body problem. The two curves for the reported extreme values of n/k_0^3 act as limiting (accumulation) curves for all practical purposes. The striking feature of Fig. 1 is that, when expressed in terms of $k_F\xi$, the behavior of the chemical potential becomes *universal* (i.e., independent of the parameter k_0 of the interaction potential), but possibly for an "intermediate" range $\pi^{-1}\lesssim k_F\xi \lesssim 2\pi$ where the normalization values ϵ_F and ϵ_0 (depending on the sign of μ) actually lose their meaning.[12] This remarkable universal behavior of μ versus $k_F\xi$ strongly suggests that $k_F\xi$ *is indeed the appropriate variable* to follow the evolution from BCS superconductivity to BE condensation.

Note in addition from Fig. 1 that μ gets pinned to (about) the normal-state value ϵ_F when $k_F\xi \gtrsim 2\pi$, and that μ drops *rather abruptly* from ϵ_F at $k_F\xi \simeq 2\pi$. In other words, Fig. 1 shows that, when the coherence length ξ equals the Fermi wavelength $\lambda_F = 2\pi/k_F$ of the electrons, the system becomes unstable against bosonization and the Fermi surface is wiped out. We expect that the instability of the Cooper-pair-based superconductivity when $k_F\xi \simeq 2\pi$, inasmuch as it is consequence of a genuine quantum-mechanical effect, should actually persist beyond the limits of validity of the procedure we have followed to establish it. In this sense, the stability criterion $k_F\xi \gtrsim 2\pi$ should be regarded as the analog for the problem at hand of the Ioffe-Regel criterion for transport in disordered systems.

It still remains to figure out how the Uemura plot for T_c versus T_F could be mapped out in terms of the variable $k_F\xi$. To this end, we remark that the variable $k_F\xi_0$ appears in the characteristic weak-limit BCS expression for T_c obtained from $k_BT_c/\Delta_{k_F} = e^\gamma/\pi$ (γ being Euler's constant) by eliminating Δ_{k_F} in favor of the Pippard coherence length ξ_0, namely,

$$k_BT_c = \frac{2e^\gamma}{\pi^2}\frac{\epsilon_F}{k_F\xi_0}\ , \tag{8}$$

k_B being Boltzmann's constant and $2e^\gamma/\pi^2 = 0.36$. The question naturally arises whether an expression like (8) could be used not only asymptotically in the weak-coupling limit, but also down to $k_F\xi \simeq 2\pi$ whenever the concept of a Fermi surface is still preserved. To answer this question, we have followed again NSR and evaluated the thermodynamic potential within the ladder approximation in the particle-particle channel for the normal state (i.e., for $T \geq T_c$). In this way, T_c and $\mu(T_c)$ have been determined versus G, or alternatively versus $k_F\xi$, for given k_0. The result is that the relation

$$k_BT_c = 0.40\frac{\epsilon_F}{k_F\xi} \tag{9}$$

holds universally for $k_F\xi \gtrsim 2\pi$, independent of k_0.[13]

We can now envisage interpreting the Uemura plot in terms of the variable $k_F\xi$, by assuming Eq. (9) to hold phenomenologically (that is, irrespective of the assumptions used to derive it) and thus superimposing on the $\log_{10}T_c$ versus $\log_{10}T_F$ plot (with $T_F = \epsilon_F/k_B$) the straight lines with $k_F\xi = \text{constant}$. The result is reported in Fig. 2 where the lines with $k_F\xi = 10$, 10^2, 10^3, 10^4, and 10^5 are drawn (full lines) together with the "instability" line $k_F\xi = 2\pi$ (broken line). We note the following features from Fig. 2: (i) *a linear trend* of $\log_{10}T_c$ versus $\log_{10}T_F$ results for any given value of $k_F\xi$; (ii) the line with $k_F\xi = 2\pi$ remarkably appears to be the natural *upper boundary* for the experimental data, suggesting that the systems near the boundary are close to a Fermi surface *instability;* (iii) what distinguishes the "exotic" from the conventional superconductors in the Uemura plot is the smaller value of $k_F\xi$ associated to the former ones (apart from a possible difference in m^*); (iv) high values of T_c result when $k_F\xi$ is "small" (~ 10) and m^* is not too large.[14]

To obtain an independent check on whether our proposal to interpret the Uemura plot is correct, one should compare the values of $k_F\xi$ associated via Eq. (9) to the various samples in the Uemura plot with *independently* measured values of $k_F\xi_{\text{exp}}$ (as obtained by critical magnetic-field measurements).[15] Preliminary checks with the experimental data available to us give indeed encouraging results for this comparison. A complete list of data on T_c, T_F, k_F, and ξ_{exp} for all samples reported in the Uemura plot is, however, required to draw a definite conclusion about the success of the comparison.

Some final comments are in order. First, we remark that intrinsic to the procedure of how the values of T_F are located in the Uemura plot is an effective angular averaging which washes out all nonspherical features of the Fermi surface. Otherwise, no meaningful connection between the Uemura plot and our expression (9) could even have been attempted. It may thus be possible that more specific criteria for instability toward bosonization, which would take into account the asymmetry of the Fermi surface (especially in reduced dimensionality), could

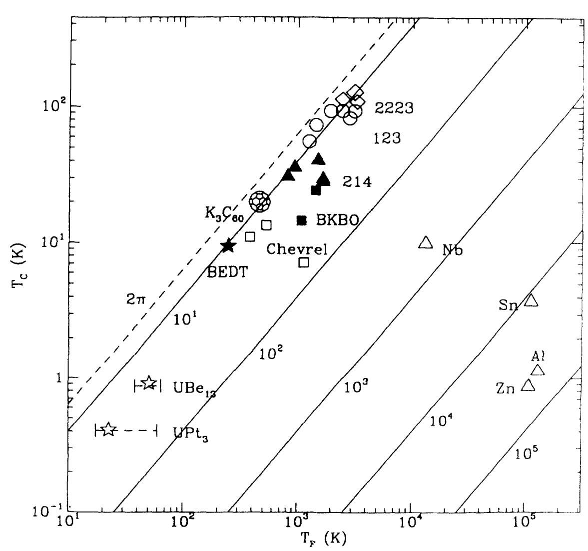

FIG. 2. Uemura plot with superimposed lines of $\log_{10}T_c$ versus $\log_{10}T_F$ according to Eq. (9) of the text for the values 2π (broken line) and 10, 10^2, 10^3, 10^4, 10^5 (full lines) of $k_F\xi$. Experimental points are reproduced from Ref. 1.

result in smaller values (than 2π) for the product of the relevant values of k and of the coherence size. Second, we emphasize that in our approach we have not taken into account any lattice effect. It is likely that the tendency toward bosonization may result in an instability of the coupled electron-lattice system (such as the onset of a charge-density wave or a structural modulation) that could actually overwhelm superconductivity. This could be possibly the reason why no physical system appears to cross the "forbidden" boundary at $k_F\xi=2\pi$ in the Uemura plot. A consequence of this sort of speculation would thus be that producing samples with even higher T_c would require one to move along the line $k_F\xi=2\pi$ in the Uemura plot.

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[1]Y. J. Uemura *et al.*, Phys. Rev. Lett. **66**, 2665 (1991); Nature **352**, 605 (1991).

[2]P. Nozières and S. Schmitt-Rink, J. Low. Temp. Phys. **59**, 195 (1985).

[3]A. J. Legget, in *Modern Trends in the Theory of Condensed Matter,* edited by A. Pekelski and J. Przystawa, Lecture Notes in Physics Vol. 115 (Springer-Verlag, Berlin, 1980), p. 13.

[4]Cf., e.g., J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964), Chap. 2.

[5]Reduction to an effective fermionic attractive potential may not be possible whenever dynamical (retardation) effects are important. For recent developments see L. Pietronero and S. Strässler, Europhys. Lett. **18**, 627 (1992).

[6]These two criteria were borne out in the original BCS suggestion to express their theoretical results in the form of *ratios* of experimentally accessible quantities, which are independent of the interaction potential and of the single-particle density of states.

[7]As in the BCS approach, we rely on a description of the normal state in terms of quasiparticles.

[8]We have eliminated at the outset the Hartree-Fock-like terms by setting $V_{\mathbf{k},\mathbf{k}}=0$ for the diagonal components. This choice will by no means invalidate our results, since it turns out that these terms are irrelevant in the parameter region of physical interest.

[9]NSR (Ref. 2) state instead that $r_0 \sim k_0^{-1}$ for $G \rightarrow \infty$. Since bosonization can be achieved only when $r_0^3 \ll n^{-1}$, NSR are able to follow the evolution from BCS to BE condensation as a function of G *only* in the "dilute limit" $n/k_0^3 \ll 1$ for the reduced (three-dimensional) density, that is, for given density n only when $k_0 \gg k_F$. This limitation has prevented NSR from connecting the two physical limits (BCS and BE) irrespective of k_0. Our finding that $r_0 \sim k_0^{-1} G^{-1/2}$, on the other hand, enables us to satisfy the bosonization condition $(n/k_0^3)/G^{3/2} \ll 1$ even in the "dense limit" $n/k_0^3 \gg 1$, provided G is large enough.

[10]Probably the most suited quantity to follow the evolution from BCS superconductivity to BE condensation is the chemical potential μ, which almost coincides with the Fermi energy in the weak-coupling limit and reduces to (half of) the lowest eigenvalue of the associated two-body problem in the strong-coupling limit. Furthermore, μ can also be the object of direct measurements: cf. G. Rietveld, N. Y. Chen, and D. van der Marel, Phys. Rev. Lett. **69**, 2578 (1992), and references quoted therein.

[11]A physical quantity directly related to $k_F\xi$ is the (average) number of electrons $\mathcal{N}_d$ within a correlation volume in d dimensions. When $d=3$ one gets $\mathcal{N}_3=(k_F\xi)^3/18\pi$, while when $d=2$ one gets $\mathcal{N}_2=(k_F\xi)^2/8$. Since in the Uemura plot three-dimensional as well as (quasi-) two-dimensional systems are treated on the same footing, we prefer to use simply $k_F\xi$ as the variable replacing G.

[12]We have verified that the universal behavior shown in Fig. 1 for $k_F\xi \gtrsim 2\pi$ and $k_F\xi \lesssim \pi^{-1}$ is *independent* of the choice of the single-particle dispersion relation $\epsilon_{\mathbf{k}}$ and of the shape of the interaction potential embodied by the function $w_{\mathbf{k}}$, provided obviously that these functions do not behave in a pathological way in **k** space.

[13]The difference between the numerical prefactors of Eqs. (8) and (9) is due to the difference between our definition (7) for ξ and the Pippard coherence length ξ_0. We have also verified that Eq. (9) holds for $k_F\xi \gtrsim 2\pi$ both in three and in two dimensions. This is a requisite to apply Eq. (9) to the Uemura plot, since the data there reported pertain to fully three-dimensional as well as to (quasi-) two-dimensional systems.

[14]It is interesting to apply Eq. (9) also to superfluid ^{3}He, for which $k_F=0.78$ Å^{-1}, $m^*/m=2.76$ (where m is the bare mass of the ^{3}He atom), and $\xi \approx 200$ Å. Equation (9) then provides for T_c the value 4.5 mK, which compares reasonably well with the experimental value (2.6 mK) if we consider the fact that Eq. (9) applies to s-wave pairing while the pairing in ^{3}He is known to be p-wave. It is thus essentially the large value of $k_F\xi$ (≈ 160) which accounts for the three orders of magnitude difference between the superfluid temperature of ^{3}He and ^{4}He. One should also mention in this context that common features between ^{3}He and heavy-fermion systems have already been pointed out by R. Tournier *et al.*, J. Magn. Magn. Mater. **76-77**, 552 (1988) (see especially their Fig. 6).

[15]When comparing the values of the coherence length ξ obtained by our definition (7) with the experimental values, one should be aware of the fact that different numerical factors (of order unity) plague alternative definitions of the (zero-temperature) coherence length. The relation between our ξ and the experimentally determined $\xi_{\rm exp}$ can be determined by relating $\xi_{\rm exp}$ to the Pippard coherence length ξ_0, in the limit when a microscopic derivation of the Ginzburg-Landau equation from BCS is justified. From the clean-limit expression $\xi_{\rm exp}=0.87\xi_0$ [cf. E. Helfand and N. R. Werthamer, Phys. Rev. **147**, 288 (1966), and references quoted therein] we then obtain $\xi \approx 1.25\xi_{\rm exp}$, which we assume to apply for *all* materials reported in the Uemura plot.