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Editors' Suggestion

Magic Gap Ratio for Optimally Robust Fermionic Condensation and Its Implications for High-T_c Superconductivity

N. Harrison and M. K. Chan
National High Magnetic Field Laboratory, Los Alamos National Laboratory,
Los Alamos, New Mexico 87545, USA

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Bardeen-Schrieffer-Cooper (BCS) and Bose-Einstein condensation (BEC) occur at opposite limits of a continuum of pairing interaction strength between fermions. A crossover between these limits is readily observed in a cold atomic Fermi gas. Whether it occurs in other systems such as the high temperature superconducting cuprates has remained an open question. We uncover here unambiguous evidence for a BCS-BEC crossover in the cuprates by identifying a universal magic gap ratio $2\Delta/k_BT_c \approx 6.5$ (where Δ is the pairing gap and T_c is the transition temperature) at which paired fermion condensates become optimally robust. At this gap ratio, corresponding to the unitary point in a cold atomic Fermi gas, the measured condensate fraction N_0 and the height of the jump $\delta\gamma(T_c)$ in the coefficient γ of the fermionic specific heat at T_c are strongly peaked. In the cuprates, $\delta\gamma(T_c)$ is peaked at this gap ratio when Δ corresponds to the antinodal spectroscopic gap, thus reinforcing its interpretation as the pairing gap. We find the peak in $\delta\gamma(T_c)$ also to coincide with a normal state maximum in γ, which is indicative of a pairing fluctuation pseudogap above T_c.

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A crossover in the pairing interactions between the weak coupling Bardeen-Schrieffer-Cooper (BCS) [1] and the strong coupling Bose-Einstein condensation (BEC) [2,3] limits was proposed in the high transition temperature T_c superconducting cuprates soon after their discovery [4–8]. On the BCS side, pairing takes place at the Fermi surface below T_c as in a conventional superconductor, whereas on the BEC side, fermions pair up to produce bosons whose subsequent condensation at T_c is determined by the phase stiffness of the superfluid. Whereas the cuprates provided the motivation for much of the early theoretical work on the BCS-BEC crossover, today it is in a cold atomic Fermi gas [9,10] where this phenomenon is well established. The relative simplicity of a cold atomic Fermi gas, consisting of pairing interactions tuned via a Feshbach resonance in an otherwise weakly interacting Fermi gas, has made it the ideal paradigm for cementing [11–13] our theoretical understanding of condensation in the crossover region [8,14]. Yet the question of whether such a crossover occurs in other paired fermion systems such as the cuprates has remained. The other proposed BCS-BEC crossover candidates include nuclear matter, quark-gluon plasmas, iron-based superconductors, and twisted graphene [15–21].

While various experiments are suggestive of a non-BCS pairing scenario in the cuprates [22–28], uncertainty has surrounded the question of whether T_c is a sufficiently large fraction of the Fermi temperature T_F for a BCS-BEC crossover to be viable [18]. For example, electronic band theory predicts a ratio $T_c/T_F \sim 10^{-2}$ that is clearly too small for a BCS-BEC crossover to occur [10]. However, thermodynamic measurements, including magnetic quantum oscillations, have revealed strongly renormalized quasiparticle effective masses [29]. It can be argued on the basis of such measurements that the ratio is close to that $T_c/T_F = 1/8$ required to be in the BCS-BEC crossover regime of a two-dimensional superconductor [18,29]. Yet, given the increased effective mass renormalizations at low temperatures [57,58] and various poorly understood phenomena such as the Fermi surface reconstruction [59,60] and "Fermi arcs" [29,61], it is unclear whether the parabolic band approximation upon which T_F estimates are based [18] is valid in the cuprates.

Studies aiming to address the question of whether a BCS-BEC crossover occurs in the cuprates [15,16,64] have instead focused on the pseudogap [65], which is a partial gap in the fermionic density of states above T_c. In a cold atomic Fermi gas, a pseudogap is reported to develop in the BEC-BCS crossover region [66–69], and is unambiguously the result of normal state pairing correlations [15,16,64,69–72]. In the cuprates, the pseudogap is maximal in the antinodal region of momentum-space where the d-wave pairing gap is maximal [65]. But while pairing has been proposed as the origin of the pseudogap in the cuprates [15,64,65,73], antiferromagnetic correlations and unconventional broken symmetry phases have also been proposed to produce a pseudogap [74–79].

In this Letter, we show that the key to establishing a universal thermodynamic signature of the BCS-BEC crossover, is the identification of a magic gap ratio [80,81] $2\Delta/k_BT_c \approx 6.5$ at which paired fermion condensates become optimally robust [16]; throughout, we use Δ to refer to the magnitude of the pairing gap at low T [14,68,82]. At this gap ratio, corresponding to the unitary point in a cold atomic Fermi gas, experimental indicators of a robust condensate exhibit a sharp peak. These include the condensate fraction N_0 and the height of the jump $\delta\gamma(T_c)$ in the fermionic (or electronic) contribution $C = \gamma T$ to the specific heat at T_c [see schematic in Fig. 1(a)]. In the cuprates, we find $\delta\gamma(T_c)$ to be peaked at the magic gap ratio when Δ corresponds to the antinodal gap [83]. Reinforcing its interpretation as the pairing gap [15,64,65,73], we find (i) nearly identical asymmetric line shapes of $\delta\gamma(T_c)$ versus $2\Delta/k_BT_c$ in the cuprates as for the unitary regime of a Fermi gas and (ii) coincidence of the peak in $\delta\gamma(T_c)$ with a normal state maximum in γ. The latter, along with an accompanying maximum in the spin susceptibility χ, can be understood as a signature of normal state pair amplitude fluctuations.

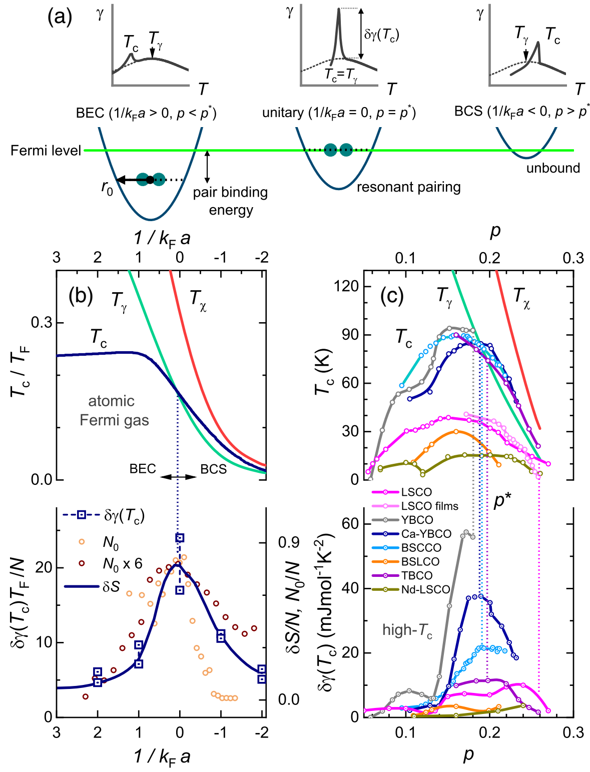

FIG. 1. (a) Schematic $\gamma(T)$ with (solid lines) and without (dotted lines) a phase transition. Because of the transition, the normal state maximum is visible only when $T_\gamma > T_c$ (or $1/k_Fa > 0$ or $p < p^*$). Also shown is a schematic of resonant pairing, occurring when the bound state energy coincides with the Fermi level [29], producing a sharp peak in $\delta\gamma(T_c)$. (b) Unitary regime of a Fermi gas [8,14]. Upper panel: T_c from Ref. [14] and $T_\gamma = 2\Delta/6.5k_B$ and $T_\chi = 2\Delta/3k_B$ (using Δ at the lowest T from Ref. [14]). Lower panel: $\delta\gamma(T_c)$ (lower and upper bound estimates extracted [29] from $S(T)$ in Fig. 5 of Ref. [14]), δS [from Fig. 6 of Ref. [14]; this closely follows $\delta\gamma(T_c)$, providing a guide to the eye], and N_0 (brown [13] and yellow [12] circles). (c) Cuprates. Upper panel: $T_c(p)$ [58,84–90,94] and T_γ and T_χ from Fig. 2(d). Lower panel: $\delta\gamma(T_c)$; spline fits connect points. In (b) and (c), dotted lines indicate $p = p^*$ (for each cuprate family [90]) and $1/k_Fa = 0$, at which $T_\gamma = T_c$, coinciding approximately with peaks in $\delta\gamma(T_c)$.

In the unitary regime of a Fermi gas, corresponding to $1 \gtrsim 1/k_Fa \gtrsim -1$ in Figs. 1(a) and 1(b), continuous tuning of the pairing interactions through the crossover occurs by way of the dimensionless parameter $1/k_Fa$ [95], where a is the pair scattering length and k_F is the Fermi radius. The BCS side [1] corresponds to $k_Fa < 0$, while the BEC side corresponds to $k_Fa > 0$. The divergence in the elastic scattering cross section at $1/k_Fa = 0$, which defines the location of the unitary point, causes the condensate to become optimally robust. This leads to peaks in $\delta\gamma(T_c)$ and in the entropy change δS accompanying condensation at T_c [14,29] [see Fig. 1(b)]. An optimally robust condensate is confirmed experimentally by the observation of a peak in N_0 as a function of $1/k_Fa$ [see Fig. 1(b)] [12,13,29] and a maximally large $\delta\gamma(T_c)$ [11,96], which also occurs at the value of T_c predicted by theory [14].

Turning to the cuprates in Fig. 1, the measured $\delta\gamma(T_c)$ changes by as much as a factor of ~30 in YBCO [58,84–90]. This change is far larger than the variations in $\delta\gamma(T_c)$ that are ordinarily explained by Eliashberg theory in regular BCS systems [29,80,81], or have been predicted in various strongly coupled pairing models of the cuprates [97–99]. The $\delta\gamma(T_c)$ curves do, however, exhibit maxima as a function of p resembling the behavior as a function of $1/k_Fa$ in the unitary regime of a Fermi gas in Fig. 1(b).

The similar behavior of the cuprates to the unitary regime of a Fermi gas becomes clear once the data from Figs. 1(b) and 1(c) are replotted on the same $2\Delta/k_BT_c$ axis in Fig. 2(a). While $2\Delta/k_BT_c$ is not a tuning parameter, it has the advantage in that it can be determined in both systems. In a cold atomic Fermi gas, there exists a direct correspondence between $1/k_Fa$ and $2\Delta/k_BT_c$ [14,100] [see Fig. 2(b)]. Studies of the unitary regime differ on the precise values of Δ and T_c at the unitary point [4,7–10,15,16,64,101]. However, they are found to be consistent with respect to the ratio $2\Delta/k_BT_c = 6.5 \pm 0.2$ [29] (see for example Fig. 9 of Ref. [100] and Table 1 of Ref. [16]), indicating this magic gap ratio to be a robust property of such a point.

Various noncuprate superconductors, including classic BCS [80] and iron-based systems [81], while spanning comparatively limited ranges in $2\Delta/k_BT_c$, are found to exhibit trends in $\delta\gamma(T_c)/\bar{\gamma}$ versus $2\Delta/k_BT_c$ consistent with Fig. 2(a) [29]. In these systems, dividing by an assumed constant Sommerfeld coefficient $\bar{\gamma}$ enables universal trends in $\delta\gamma(T_c)$ to be established [29] for materials with different electronic structures. The iron-based superconductors with the highest $\delta\gamma(T_c)/\bar{\gamma}$ values are found to have gap ratios consistent with the magic value. Of these, iron selenide has also recently been reported to exhibit a BCS-BEC

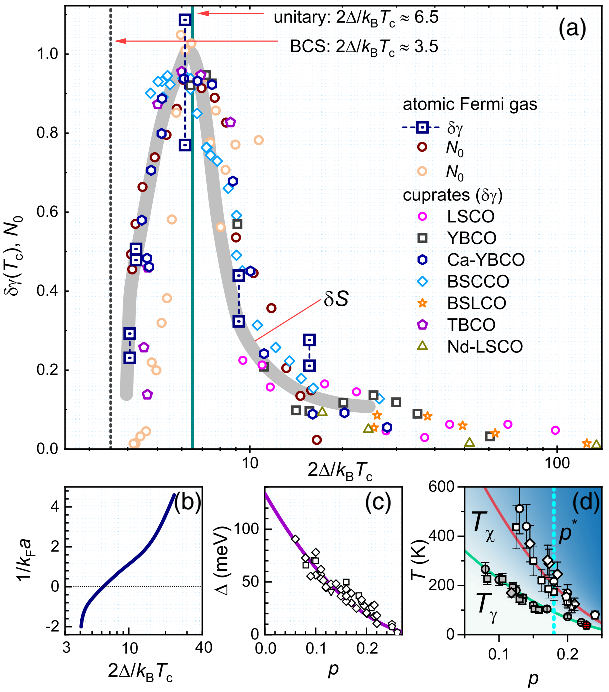

FIG. 2. (a) $\delta\gamma(T_c)$, N_0, and δS [rescaled to unity from Figs. 1(b) and 1(c)] versus $2\Delta/k_BT_c$. (b) $1/k_Fa$ versus $2\Delta/k_BT_c$ [14]. (c) Spectroscopic and thermal antinodal gap measurements [73,102,103]. (d) Maxima in γ (grey symbols) and χ (white symbols) from the raw data [29]. T_γ (green line) is a polynomial fit to the grey symbols [104], from which we obtain $\Delta = 6.5k_BT_\gamma/2$ [i.e., the purple line in (d)] and $T_\chi = 2\Delta/3k_B$ (red line). Symbol shapes identify the cuprate family in (a), (c), and (d). The down triangle in (c) refers to HBCO [90].

crossover [20,21]—albeit without accompanying measurements of $\delta\gamma(T_c)$. A similar gap ratio at unitarity is further reported in gated layered superconductors [105].

The asymmetric line shape in Fig. 2(a) can be understood to result from the fact that the gap ratio has a hard cutoff on the left-hand side at a value similar to that ≈3.5 of an ideal BCS superconductor [1], while there is no cutoff on the BEC side [4,7–10,15,16,64]. On the BCS side, $\delta\gamma(T_c)$ increases with $2\Delta/k_BT_c$ similarly to that in Eliashberg theory [29,80], while on the BEC side, T_c and consequently $\delta\gamma(T_c)$ are limited by the phase stiffness of the condensate [23]. We find precisely this line shape in the cuprates when Δ [purple line in Fig. 2(c)] [29,104] corresponds to the antinodal gap dominating spectroscopic and thermodynamic measurements [symbols in Fig. 2(c)] [73,102,103]. The same asymmetric behavior is displayed for multiple cuprate families [58,84–89]. On averaging the values of $2\Delta/k_BT_c$ in Fig. 2(a) at which $\delta\gamma(T_c)$ is peaked [near $p \approx 0.2$ in Fig. 1(c)] for the higher T_c cuprates (YBCO, Ca-YBCO, BSCCO, and TBCO [90]), we obtain $2\Delta/k_BT_c = 6.4 \pm 0.3$, which is the same within experimental uncertainty as for a unitary Fermi gas. Validity of the universal magic gap ratio is therefore strongly suggested in the cuprates.

The association of Δ with the antinodal gap in the cuprates is reinforced by thermodynamic evidence for pairing correlations in the normal state. In the cuprates, normal state pair amplitude fluctuations associated with the pseudogap have been proposed to account for maxima in γ and χ as a function of T [97–99,106]. Pair amplitude fluctuations in the unitary and BEC regimes of a Fermi gas also produce normal state maxima in γ (or $C = \gamma T$) [29,107] and χ [71,72,108,109]. Figure 3 shows that on plotting γ [87,110] and χ [87,91,92,111–116] versus $2\Delta/k_BT$, maxima in γ and χ emerge as ubiquitous properties of the normal state (in the cuprates, the shape of χ versus T is provided by magnetic susceptibility χ_m and nuclear magnetic resonance Knight shift K measurements [117]). The model γ and χ curves (black and grey in Fig. 3) produced by an excitation gap of width Δ [29,83] exhibit maxima at $T_\gamma \approx 2\Delta/6.5k_B$ and $T_\chi \approx 2\Delta/3k_B$. A pairing pseudogap [118] is strongly suggested in the cuprates by the consistency of the observed maxima in Fig. 3 with T_γ and T_χ. In fact, we find overall consistency between each of the $\Delta(p)$, $T_\gamma(p)$, and $T_\chi(p)$ curves and the experimental data points for the antinodal gap and maxima in γ and χ [29] in Figs. 2(c) and 2(d). Thermodynamic and spectroscopic measurements can therefore both be understood in terms of a $\Delta(p)$ that is approximately the same for all cuprate families, regardless of their optimal T_c.

A direct association of the normal state maxima with pairing amplitude fluctuations is strongly suggested by the alignment of the maxima in γ with the peaks in $\delta\gamma(T_c)$ when γ and $\delta\gamma(T_c)$ are, respectively, plotted versus $2\Delta/k_BT$ and $2\Delta/k_BT_c$ in Fig. 3(a). Since $2\Delta/k_BT$ and $2\Delta/k_BT_c$ are both scaled by Δ, the alignments of γ and $\delta\gamma(T_c)$ are independent of any experimental uncertainties in the functional form of $\Delta(p)$ [104]. We find the alignments to originate from $\delta\gamma(T_c)$ being peaked close to the points of intersection of T_c with T_γ [see Figs. 1(b) and 1(c)], corresponding to $1/k_Fa = 0$ (i.e., the unitary point) in a unitary Fermi gas and a characteristic doping $p = p^*$ in the cuprates.

In the unitary regime of a Fermi gas, $\delta\gamma(T_c)$ exhibiting a strong peak at $T_c = T_\gamma$ can be understood as a consequence of the heavily broadened pseudogap transitioning into a regular pairing gap [118] as long range phase coherence is established below T_c [119,120]. The entropy change contributing to $\delta\gamma(T_c)$ is naturally largest when T_c coincides with the maximum in γ resulting from excitations across Δ. This is therefore suggested also to occur in the cuprates at $T_c = T_\gamma$ [29]. $\delta\gamma(T_c)$ exhibiting a strong peak at $T_c = T_\gamma$ can also be understood as a consequence of the normal state entropy S_n at T_c (in addition to δS) exhibiting a maximum (as a function of $1/k_Fa$) close to this point, owing to this region of the normal state consisting of a maximally disordered mixture of a bosonic and fermionic degrees of freedom [14,107]. At $T > T_c$, a peak in $S_n(1/k_Fa)$ is also seen to extend vertically in T at the

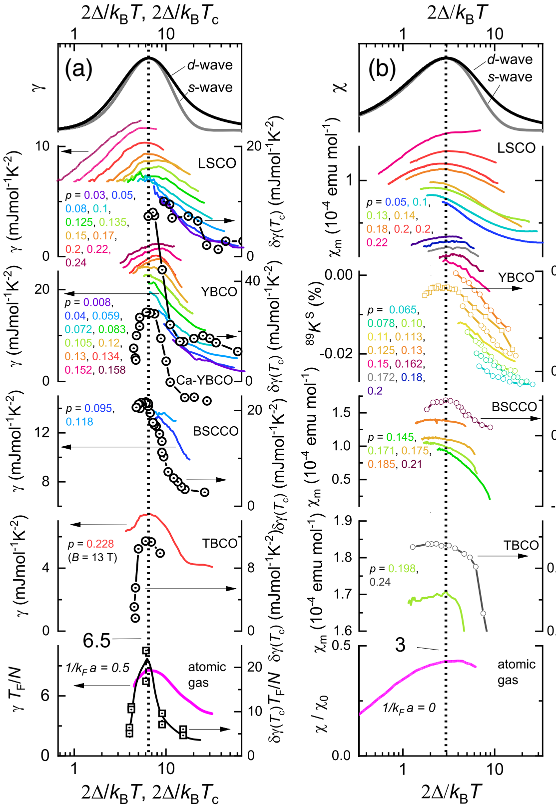

FIG. 3. (a) Left-hand axes: γ for $T > T_c$ [29,37,64,84,87,110] (colored lines; including 2% Zn substitution for the highest 2 YBCO dopings [110]) versus $2\Delta/k_BT$; p and $1/k_Fa$ values are indicated throughout. Right-hand axes: $\delta\gamma(T_c)$ at T_c versus $2\Delta/k_BT_c$ for the cuprates [58,84–88] (center-dot circles; shifted by ±10 mJ mol^{-1} K^{-2} for YBCO and Ca-YBCO) and a Fermi gas from Fig. 1 (center-dot squares). (b) χ_m [87,92,112,113], K [91,111,114–116] and χ [108] versus $2\Delta/k_BT$ [117]. Included are γ and χ for model d- (black) and s-wave (grey) gaps of magnitude Δ [29]. Some YBCO K curves are shifted vertically for clarity and spline fits connect coarsely spaced points. Dotted lines indicate $2\Delta/k_BT_c = 6.5$ and $2\Delta/k_BT = 3$.

unitary point [9,14], with the loss of fermion degrees of freedom at $1/k_Fa > 0$ leading to a sharp drop S_n on the BEC side of the phase diagram. An examination of S_n in several cuprates [121] reveals that this too exhibits a sharp peak that extends vertically in T near p^*, accompanied by a drop in S_n at $p < p^*$.

One consequence of $\delta\gamma(T_c)$ being peaked close to the point of intersection of T_γ and T_c is that p^* is distinct from the hole doping $p \approx 0.16$ at which T_c is optimal. In fact, p^* moves towards the upper end of the superconducting dome as the optimal T_c is reduced, and appears to be accompanied by a strong suppression of the overall peak height of $\delta\gamma(T_c)$. In LSCO, for example, an extrapolation of T_γ in Fig. 1(c) suggests that $p^* \approx 0.26 \pm 0.03$, which is consistent with the higher value of $p = 0.23 \pm 0.1$ (compared, e.g., to YBCO) at which $\delta\gamma(T_c)$ is peaked [29] and the higher value of $p = 0.24 \pm 0.01$ (compared to Ca-YBCO) at which S_n is peaked [121]. In LSCO films, by contrast, T_c lies significantly below T_γ in Fig. 1(c), suggesting that they do not exhibit a crossover into the BCS regime, as has also been suggested on the basis of the superfluid density measurements [29,94]. The tiny p-dependent $\delta\gamma(T_c)$ in Nd-LSCO, meanwhile, suggests that its peak value occurs at higher dopings than have been accessed experimentally [58,122].

Given the prior reports of quantum criticality in the cuprates at similar hole dopings to p^* [123–125], one intriguing possibility is that the BCS-BEC crossover and quantum criticality share a common origin. Indeed, some of the reported phenomenology of quantum criticality in the cuprates bears similarities to that of the unitary regime of a Fermi gas [126]. This includes Plankian dissipation and scale invariance [123–128], and a minimum in the pair coherence length [17,129,130] inferred from the maximum in the superconducting upper critical magnetic field [29,57,93,131]. It should be noted, however, that thermodynamic evidence for quantum criticality in the form of a sharply increasing γ or an upturn in the effective mass, has thus far only been reported at low temperatures ($T \ll 10$ K) [57,58], and has yet to be accompanied by evidence for a divergence in the correlation length of a broken symmetry phase [29].

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[29] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.129.017001, which discusses data pertaining to $\delta\gamma(T_c)$ in conventional BCS and Fe- and Ni-based superconductors; information about cuprate hole dopings p used in constructing graphs; raw γ and χ data including the locations of T_γ and T_χ; Knight shift data on BLSCO; a discussion pertaining to prior modeling of $\delta\gamma(T_c)$ versus p by Chen *et al.*; details concerning cold atomic gas data used; estimates of the gap ratio at the unitary point of a cold atomic gas; a discussion of the thermodynamics of maxima in γ and χ; coherence length estimates in various cuprates based on H_{c2}; estimates of the Fermi energy in the cuprates; a discussion of the higher values of p^* in cuprates with lower T_c's; the origin of "Fermi arcs"; the T dependence of the antinodal gap; lifetime effects on N_0; problems with a smaller gap option for Δ in the cuprates; a hidden maximum in the heat capacity of a cold atomic Fermi gas; reports of peaks in $\delta\gamma(T_c)$, γ and m^* associated with quantum criticality, and includes Refs. [30–56].

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