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A universal relationship between magnetic resonance and superconducting gap in unconventional superconductors

G. Yu[1], Y. Li[1], E. M. Motoyama[1] and M. Greven[2,3]*

Superconductivity involves the formation of electron pairs (Cooper pairs) and their condensation into a macroscopic quantum state. In conventional superconductors, such as $\mathbf{Nb_3Ge}$ and elemental Hg, weakly interacting electrons pair through the electron–phonon interaction. In contrast, unconventional superconductivity occurs in correlated-electron materials in which electronic interactions are significant and the pairing mechanism may not be phononic. In the cuprates, the superconductivity arises on doping charge carriers into the copper-oxygen layers of antiferromagnetic Mott insulators[1]. Other examples of unconventional superconductors are the heavy-fermion compounds, which are metals with coupled conduction and localized *f*-shell electrons[2], and the recently discovered iron–arsenide superconductors[3]. These unconventional superconductors show a magnetic resonance, a prominent collective spin-1 excitation mode in the superconducting state[4–8]. Here we demonstrate the existence of a universal linear relation, $E_r \propto 2\Delta$, between the magnetic resonance energy (E_r) and the superconducting pairing gap (Δ), which spans two orders of magnitude in energy. This relationship is valid for the three different classes of unconventional superconductors, which range from being close to the Mott-insulating limit to being on the border of itinerant magnetism. As the common excitonic picture of the resonance has not led to such universality, our observation suggests a much deeper connection between antiferromagnetic fluctuations and unconventional superconductivity.

The resonance is a widely discussed feature in the magnetic excitation spectrum of the cuprates. It is a collective mode at a well-defined energy that appears in the superconducting (SC) state at the two-dimensional antiferromagnetic wavevector (π/a, π/a; refs 4, 5). Early inelastic neutron scattering experiments for $YBa_2Cu_3O_{6+\delta}$ (YBCO) revealed a single resonance mode. However, the crystal structure of YBCO contains two nearby Cu–O layers per unit cell, and later experiments revealed at higher energy an even-parity resonance that differs from the previously observed odd-parity mode in its symmetry with respect to the exchange of the two layers[9–11]. The resonance is also observed in neutron scattering measurements of other hole-doped cuprates: double-layer $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212; refs 12, 13), and the single-layer systems $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201; ref. 14) and $HgBa_2CuO_{4+\delta}$ (Hg1201; ref. 15), which show only one resonance mode. Investigation of $Pr_{1-x}LaCe_xCuO_{4+\delta}$ (PLCCO; ref. 16) and $Nd_{2-x}Ce_xCuO_{4+\delta}$ (NCCO; ref. 17) suggests that the resonance exists in the electron-doped cuprates as well. Magnetic peaks at the antiferromagnetic zone centre were also found in the SC state of the heavy-fermion superconductors UPd_2Al_3 (ref. 6) and $CeCoIn_5$ (ref. 7), as well as in the iron arsenides $Ba_{1-x}K_xFe_2As_2$ (ref. 8; hole doped) and $BaFe_{2-x}Co_xAs_2$ (ref. 18; electron doped), indicating that the resonance is a fairly universal feature in the magnetic spectra of unconventional superconductors.

For the cuprates, the mode energy E_r of the resonance is commonly compared with the superconducting transition temperature T_c. Figure 1 shows that the linear correlation $E_r = 5$–$6k_BT_c$ is approximately satisfied for the odd-parity mode of the double-layer compounds YBCO and Bi2212, for the resonance in Tl2201 and even for the characteristic energy of the (momentum-integrated) local susceptibility in $La_{2-x}Sr_xCuO_4$ (LSCO; refs 19, 20), in which no resonance occurs at $(\pi/a, \pi/a)$. However, recent experiments reveal a violation of the proportionality between E_r and T_c for single-layer Hg1201 (ref. 15) and for electron-doped NCCO (ref. 17). Furthermore, below optimal doping (hole concentration $p < 0.16$), the odd and even resonance energies of the double-layer compounds differ significantly. The average of the two is considerably larger than $6\,k_BT_c$ for the most underdoped YBCO samples in which both modes have been studied[11,21] (Fig. 1).

The characteristic energy scale of superconductivity is the SC gap Δ in the low-energy single-particle response. As seen from Fig. 2, our analysis shows that E_r is universally related to Δ rather than to T_c. We note that for the hole-doped cuprates the determination of the SC gap is complicated by the existence of multiple energy scales, the deviation of the gap function from a pure *d*-wave form and spatial inhomogeneity of the gap energy. From photoemission measurements, the SC gap is associated with the gap in the low-energy charge excitations near the nodal region in momentum space, whereas a distinct 'pseudogap' is observed in the antinodal region[22]. In the underdoped regime, we estimate the SC gap maximum Δ from the nodal gap Δ_0 determined in recent photoemission experiments. Above optimal doping, we estimate Δ from tunnelling, thermal conductivity and the spatially averaged gap determined in recent scanning tunnelling microscopy measurements. The details of our estimation of Δ in the hole-doped cuprates are found in the Supplementary Information. For the iron arsenides, two SC gaps are observed on different pieces of Fermi surface. We consider the larger of the two, which is associated with nesting bands that are thought to be responsible for the magnetic resonance. For all of the other systems, the SC gap is obtained directly from the literature.

We estimate $E_r/2\Delta = 0.64(4)$ from a fit to the combined result (Fig. 2), which spans two orders of magnitude in energy. A possible connection between the resonance energy and the SC gap has been previously suggested on the basis of observations for the heavy-fermion compound $CeCoIn_5$ (ref. 7), and through

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[1]Department of Physics, Stanford University, Stanford, California 94305, USA, [2]School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA, [3]Department of Applied Physics, Stanford University, Stanford, California 94305, USA. *e-mail: greven@physics.umn.edu.

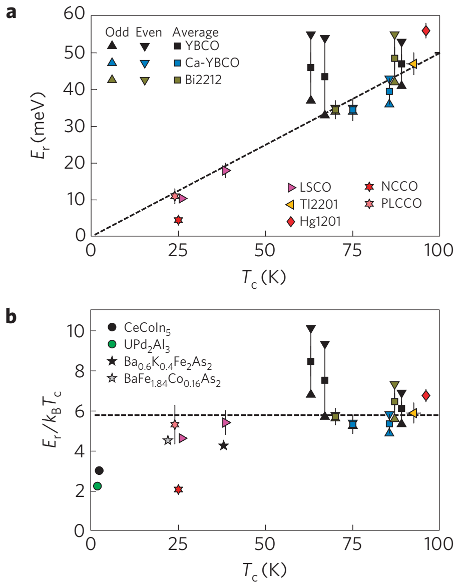

Figure 1 | Relation between the resonance energy $E_{\rm r}$ and the transition temperature $T_{\rm c}$. a, $E_{\rm r}$ versus $T_{\rm c}$ for cuprates. For the double-layer hole-doped cuprates (YBCO, Ca-doped YBCO and Bi2212), up (down) triangles indicate odd (even) resonance modes, whereas squares represent the average of the two mode energies[9-11,13,21]. For LSCO, the resonance is absent and the characteristic energy shown is the maximum in the local magnetic susceptibility, defined as the integral of $\chi''(\mathbf{Q},\omega)$ over the Brillouin zone[19,20]. The interpretation of neutron data for the magnetic resonance in the electron-doped cuprates PLCCO (ref. 16) and NCCO (ref. 17) is discussed in the Supplementary Information. The dashed line indicates $E_{\rm r} = 5.8\, k_{\rm B}T_{\rm c}$. The results for Hg1201 (ref. 15), NCCO (ref. 17) and the most underdoped YBCO samples[11,21] deviate strongly from this relationship. Supplementary Fig. S1 shows the estimated doping dependence of $E_{\rm r}$ for the hole-doped cuprates, including additional results for the odd-parity mode. **b**, The ratio $E_{\rm r}/T_{\rm c}$ versus $T_{\rm c}$ for all three types of unconventional superconductor. In addition to the results in **a**, this figure includes those for the heavy-fermion superconductors UPd$_2$Al$_3$ (ref. 6) and CeCoIn$_5$ (ref. 7) as well as for the iron arsenides Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ (ref. 8) and BaFe$_{1.84}$Co$_{0.16}$As$_2$ (ref. 18). All four show a rather small value of $E_{\rm r}/T_{\rm c}$. The resonance energy and $T_{\rm c}$ values are summarized in Supplementary Table S1. Whenever the error bar can be estimated (see Supplementary Information), it represents one-sigma confidence.

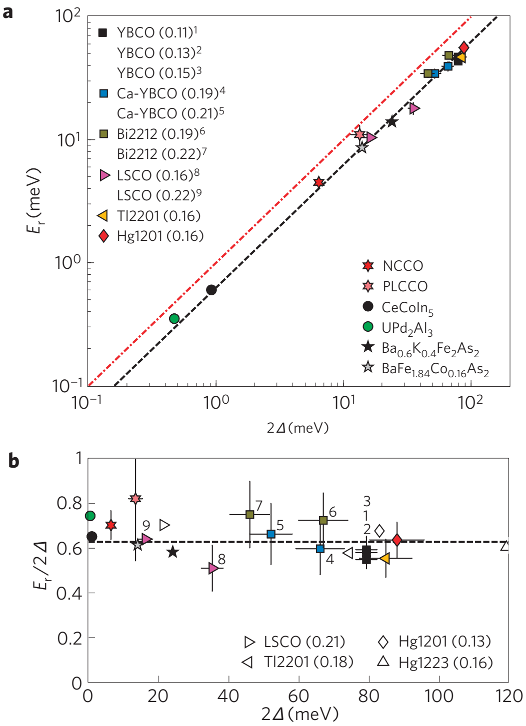

Figure 2 | Universal relationship between the resonance energy $E_{\rm r}$ and the SC gap Δ. a, The resonance energy $E_{\rm r}$ versus the pair-breaking energy 2Δ (twice the SC gap). $E_{\rm r}$ of the double-layer cuprates is represented by the average of the even- and odd-parity mode energies. The black dashed line is a fit to all of the data: $E_{\rm r}/2\Delta = 0.64(4)$. Unlike the case in Fig. 1, the results for Hg1201 (ref. 15), NCCO (ref. 17), and the average mode energy of the most underdoped YBCO samples[11,21] all follow this relationship. As a reference, the red dot-dashed line represents $E_{\rm r} = 2\Delta$. Supplementary Fig. S2 shows the resonance energy as a function of 2Δ on a linear scale. **b**, The ratio of $E_{\rm r}/2\Delta$ versus 2Δ. The dashed horizontal line indicates the value $E_{\rm r}/2\Delta = 0.64$. The open symbols indicate the maximum in the bosonic spectral function ($E_{\rm opt}$) extracted from optical spectroscopy for the single-layer cuprates LSCO, Tl2201 and Hg1201, and for triple-layer Hg1223 (ref. 27). For the hole-doped cuprates, estimated hole concentrations are shown in parentheses. The 2Δ values and hole concentrations are obtained as discussed in the Supplementary Information. The characteristic energy and SC gap values are summarized in Supplementary Table S1 (neutron scattering results) and Supplementary Table S2 (optical conductivity results). Whenever the error bar can be estimated (see Supplementary Information), it represents one-sigma confidence.

a comparison of the odd mode in the double-layer cuprates with the coherent SC gap observed by Andreev reflection[23]. Here we emphasize the universality of this connection across various classes of the unconventional superconductors and, in particular, we arrive at a consistent picture for the entire family of cuprates. For the double-layer systems, which show two resonance modes, we use the mean value of $E_{\rm r}$ in the analysis, rather than only the odd-parity mode.

The magnetic resonance seems to be associated with a SC gap function that undergoes a sign change. This is naturally the case in a Fermi-liquid picture for the d-wave cuprates, because the singularity of the magnetic susceptibility owing to the coherence factor appears at momenta $\mathbf{Q} = (\pi/a, \pi/a)$ that satisfy $\Delta(\mathbf{q}+\mathbf{Q}) = -\Delta(\mathbf{q})$ (ref. 5). We note that, although weak-coupling theory might be appropriate for the very overdoped part of the cuprate phase diagram, it is inadequate at lower doping[1]. The heavy-fermion superconductors considered here also seem to show a d-wave order parameter[2]. On the other hand, the SC gap of the iron arsenides might be nodeless. Nevertheless, the existence of the resonance may be accounted for by the possible antiphase correlation between nesting hole and electron pockets responsible for the magnetic excitations[8]. This is further discussed in the Supplementary Information.

The significance of the linear scaling relation demonstrated in Fig. 2 lies in its simplicity and its universality among different types of unconventional superconductor, which has not been predicted theoretically. In some theoretical models, the resonance is considered to be a π mode, due to staggered d-wave particle–particle charge ±2 excitations, or a magnetic plasmon, as a result of mixing between the spin and charge channels[24,25]. The most widely considered theoretical model views the resonance as a spin exciton: a spin-1 particle–hole excitation with momentum $\mathbf{Q} = (\pi/a, \pi/a)$ that is bound at an energy below the pair-breaking energy (that is, $E_{\rm r} < 2\Delta$) (ref. 5). The energy difference $2\Delta - E_{\rm r}$ depends on the specific interactions (for example, spin or charge correlations) stabilizing the bound particle–hole pair and is expected to differ among different classes of superconductors, and for different doping regimes of the hole-doped cuprates. Applied to the hole-doped cuprates, random-phase-approximation approaches to the excitonic picture generally predict the ratio $E_r/2\Delta$ to be near one for the very overdoped regime, and to significantly decrease with decreasing hole concentration, rather than to take on a universal value[5]. Such approaches assume weak coupling and are thus expected to break down in the underdoped regime. A recent strong-coupling calculation for the Hubbard model near optimal doping predicts a significant increase of the resonance energy E_r with doping[26], consistent with the behaviour of the odd-parity mode in underdoped YBCO, but inconsistent with the doping independence for the average mode energy of YBCO.

One explanation for the unconventional superconductivity is that it is mediated by magnetic fluctuations. In that case, the SC energy scale Δ would naturally be expected to follow from magnetic energy scales. Such a model implies that the electron–boson spectral function, which contains information about the 'pairing glue' for superconductivity, should bear a clear signature of the magnetic spectra. For the cuprates, estimates of the electron–boson spectral function have been obtained by inverting optical spectra[27,28]. One approach[27] has revealed a well-defined peak in the SC state at a characteristic energy E_{opt} that agrees remarkably well with E_r (Fig. 2b), although for the double-layer compounds Bi2212 and YBCO E_{opt} seems to be dominated by the odd-parity resonance mode. For triple-layer Hg1223, for which no neutron data are yet available, $E_{\mathrm{opt}} \approx 72$ meV (ref. 27) agrees well with the scaling established in Fig. 2.

Optical spectroscopy is an inherently momentum-integrated probe, and thus the extracted electron–boson spectral function is related to a momentum integral of the magnetic susceptibility $\chi''(\mathbf{Q}, \omega)$ rather than the value at the specific momentum $\mathbf{Q} = (\pi/a, \pi/a)$. Furthermore, it has been argued that the magnetic resonance itself has relatively small spectral weight[29] and that magnetic fluctuations throughout an extended momentum region near $(\pi/a, \pi/a)$ may be relevant to superconductivity[30,31]. Thus the important magnetic energy scale might be the characteristic energy of the local susceptibility $\chi''(\omega)$. The data seem to be consistent with a scaling between the SC gap and such an energy. For example, for $\mathrm{CeCoIn_5}$ (ref. 7), the resonance is very prominent, so that the characteristic energy of $\chi''(\omega)$ is expected to be close to the value of the resonance energy. For NCCO, the characteristic energies of the peak and local susceptibilities are nearly indistinguishable as well[17]. LSCO is a particularly revealing case: although the main contribution to $\chi''(\omega)$ below 25 meV does not come from $(\pi/a, \pi/a)$, its characteristic energy does indeed scale with the SC gap (Figs 1 and 2).

The observation of a universal connection between the magnetic resonance energy and the SC gap for a wide range of materials seems to indicate that magnetic excitations play an important part in the Cooper-pair formation. To fully understand the connection between the electron–boson spectral function and the magnetic susceptibility, including the resonance, more information from inelastic neutron scattering is needed. Measurements of the doping and temperature dependence of the full magnetic susceptibility of single-layer Hg1201 and of the combined even- and odd-parity excitations in double-layer YBCO and Bi2212 are particularly desirable.

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Additional information

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