Magnon-Exchange Pairing and Superconductivity

A recent suggestion by G. Chen and W. A. Goddard (*1*) for electron pairing in high-temperature superconducting oxides reintroduced the concept of magnon exchange as a replacement for the standard Bardeen-Cooper-Schrieffer (BCS) phonon exchange (*2*). This suggestion received considerable attention because [on the basis of microscopic calculations (*1, 3*) for small clusters] it provided precise estimates of the various superconducting transition temperatures T_c in the cuprate ceramics and calculated an upper bound $T_c^{max} = 232$ K. We show that, within the Chen-Goddard mechanism, the estimates of T_c are incorrect because Chen and Goddard use an equation for T_c appropriate only for weak coupling and that their T_c^{max} is spurious, as there is no upper bound when the correct expression is used.

The Chen-Goddard calculation makes use of the weak-coupling BCS model (*2*)

$$T_c = 1.13\ T_0 e^{-1/\lambda} \qquad (1)$$

where, in the Chen-Goddard mechanism, $T_0 = |J_{dd}| = 205$ K is a Cu-Cu magnetic exchange parameter, and the dimensionless coupling constant λ is $(N_0|J_{pd}|^2)/(2\tau|J_{dd}|)$. Here N_0 is the band density of states, J_{pd} is the magnetic coupling of nearest neighbor Cu and O atoms, and $0 \leq \tau \leq 1$ measures the randomness of the neighboring Cu magnetic moments, with $\tau = 0$ representing complete randomness. Estimates of the parameters (*3*) yield λ values of $0.0705\ \tau^{-1}$ for $La_{1.85}Sr_{0.15}CuO_4$, and $0.00609\ \tau^{-1}$ for the chains of $YBa_2Cu_3O_y$ with $6.8 \leq y \leq 7$. For the sheets of $YBa_2Cu_3O_y$, Chen and Goddard use the parameters from $La_{1.85}Sr_{0.15}CuO_4$ with $\tau = 0.02$. With these values, Eq. 1 yields $T_c = 114$ K and 174 K for $\tau = 0.05$ and 0.02, respectively. For $\lambda \rightarrow \infty$, one obtains $T_c = T_c^{max} = 1.13\ |J_{dd}| = 232$ K.

McMillan (*4*) augmented the weak coupling BCS expression in Eq. 1 to include renormalization. This results in the constant prefactor changing from 1.13 to 0.69, and λ being replaced by $\lambda^* = \lambda/(1 + \lambda)$. This change, valid for $\lambda < \sim 1.5$, is significant when $\lambda \sim 1$. For the Chen-Goddard estimate of $T_c = 114$ K, $\lambda = 1.41$. Hence $\lambda^* = 0.53$ and T_c becomes 21 K. For the Chen-Goddard estimate of $T_c = 174$ K, $\lambda = 3.52$ and the McMillan equation breaks down. It is appropriate, however, to use an expression obtained either as a fit (*5*) to the Eliashberg equations or analytically (*6*), and which gives a reasonable estimate of T_c

$$T_c = a|J_{dd}|(e^{2/\lambda} - 1)^{-1/2} \qquad (2)$$

where $a = 0.25$ gives the correct McMillan limit. For $\lambda = 3.52$, $T_c = 58$ K.

The estimate of $T_c^{max} = 232$ K is a spurious result, derived from the weak coupling expression (Eq. 1). If Eq. 2 is used, at the large λ limit we recover the Allen-Dynes (*7*) limiting expression $T_c = 0.18\ |J_{dd}|\lambda^{1/2}$, and T_c has no upper bound as $\lambda \rightarrow \infty$. Within the Chen-Goddard mechanism, estimates of T_c should be changed from 114 K, 174 K, and 232 K to 21 K, 58 K, and ∞, respectively.

At this time it is generally accepted that the identity of the exchange boson for the superconducting pairing electrons in the oxides is still an open question. Phonons, excitons, plasmons, and magnons are among the candidates (*8*), and there are more. In all cases the appearance of a superconducting instability (*9*) in the original (normal) state has to compete against other, usually energetically more favorable, instabilities. For the magnon exchange mechanism the dominating instability is normally another magnetic phase, for example, ferromagnetism, spiral spin arrangements, or spin glasses.

If the superconducting state is stable in some temperature range, then a T_c^{max} may possibly exist if λ in the exponent and the prefactor $|J_{dd}|$ of Eq. 2 are both renormalized.

It is notoriously difficult to predict the existence of new superconductors and to calculate T_c, even for conventional electron-phonon coupling (*10*), because large changes in T_c are usually found for small changes in coupling. Hence the proposal by Chen and Goddard to test their theory with the use of microscopic electronic calculations of their material parameters is very attractive. However, the cluster calculations of Guo *et al.* (*3*) give at best rough estimates of the electrical parameters on the scale needed.

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Response: Cohen and Falicov's (*1*) interpretation of our reasoning (*2*) regarding the maximum achievable T_c is incorrect. The upper limit for T_c can be estimated by using the following equation (*3*)

$$T_c = \frac{\langle\omega\rangle}{1.20} \exp[-1.04(1 + \eta)/\eta] \qquad (1)$$

valid for $\eta \sim 1$, where in our case

$$\eta = \frac{\lambda N(0)(J_{pd})^2}{8\tau\beta|J_{dd}|} \qquad (2)$$

and

$$\langle\omega\rangle = \frac{2}{\eta}\int \alpha^2 F(\omega)d\omega \qquad (3)$$

The upper limit of $\langle\omega\rangle$ is $4J_{dd}$, while the upper limit of the exponential term is e^{-1}, leading to $T_c < 1.23\ |J_{dd}|$. Our calculated value of $|J_{dd}|$ ~200 K leads to $T_c < 246$ K.

For $\eta \gg 1$, the correct formula (*3*) is $T_c = 0.18\ \sqrt{\langle\omega^2\rangle\eta}$, where

$$\langle\omega^2\rangle = \frac{2}{\eta}\int a^2 F(\omega)d\omega$$

Estimates of the integral in $\langle\omega^2\rangle$ using various forms for $F(\omega)$ lead to $T_c < |J_{dd}|$.

Thus the upper limit on T_c corresponds to parameters such that η is of magnitude one and leads to

$$T_c^{max} \approx 1.23|J_{dd}| \approx 246 \text{ K}$$

In our paper (*2*) we approximated this as

$$T_c^{max} \approx J_{dd} \approx 200 \text{ K}$$

which we still consider to be a reasonable but conservative estimate.

In our analysis of the maximum T_c we presume that the values for J_{pd} and J_{dd} are constrained within tight limits by the character of the relevant orbitals in the Cu-O sheets (this leads to $|J_{dd}| \approx 200$ K and $|J_{pd}| \approx 400$ K, values that increase as the