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# Prediction of superconductivity in $Am_{2-z}Ce_zCuO_4$

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In the charge-reservoir oxygen model, $Am_{2-z}Ce_zCuO_4$ is predicted to be a *p*-type high-temperature superconductor, with $T_c$ around 24 K, provided the sample geometry is selected to minimize the effects of Am radioactive decay. The cuprate-plane model of high-temperature superconductivity cannot be sustained if $Am_{2-z}Ce_zCuO_4$ superconducts and $Cm_{2-z}Th_zCuO_4$ continues to not superconduct. [S0163-1829(99)15021-5]

## I. INTRODUCTION

The true test of a theory of superconductivity is its ability to predict new superconductors. To our knowledge, this test has been passed successfully only a very few times: by Cohen's prediction of superconductivity in $SrTiO_3$ at 1 K and in other *low-temperature* superconductors [1], and by the predictions for the high-temperature superconductors $PrBa_2Cu_3O_7$ ($T_c \approx 90\,K$),[2,3] $Gd_{1.6}Ce_{0.4}Sr_2Cu_2TiO_{10}$,[4,5] and $Pr_{1.5}Ce_{0.5}Sr_2Cu_2NbO_{10}$ ($T_c \approx 25-28\,K$).[6,7] In the last three cases, the predictions were made for compounds isostructural to materials that do superconduct, using the self-consistent bond-charge method,[8] and were based on the charge-reservoir oxygen model.[9] This model employs structural considerations primarily and admits to an incomplete knowledge of the dynamics of electrons in the superconducting state. The main difference between the charge-reservoir oxygen model and the cuprate-plane models[10] of high-temperature superconductivity is that the primary superconducting condensate is in the charge reservoirs (vicinity of the chain layers of $PrBa_2Cu_3O_7$, La-O layers of $La_{2-z}Sr_zCu_4$, or Sr-O layers of $Pr_{2-z}Ce_zSr_2Cu_2NbO_{10}$), while this condensate is in the $CuO_2$ planes for the cuprate-plane models.

However, the charge-reservoir oxygen model stands in opposition to the popular cuprate-plane picture of high-temperature superconductivity, and so its successes in having predicted three high-temperature superconductors are regarded by some proponents of cuprate-plane superconductivity as accidents. Nevertheless, three successful predictions of new superconductors cannot be ignored when the more popular cuprate-plane models taken collectively have none—especially since cuprate-plane models have been unable to explain why the high-temperature superconductors $Ba_{1-a}K_aPb_{1-b}Bi_bO_3$ ($T_c \approx 32\,K$) (Ref. 11) and Cu-doped $YSr_2RuO_6$ ($T_c \approx 82\,K$) (Ref. 12) can superconduct with no cuprate planes, and why rare-earth-site Pr in $PrBa_2Cu_3O_7$ does not adversely affect $T_c$, while Ba-site Pr (which is symmetrically placed with respect to the cuprate plane in between the Ba and rare-earth sites) destroys superconductivity.[2] Accordingly, we suggest another experimental test, one that discriminates between the charge-reservoir oxygen and cuprate-plane models: the charge-reservoir oxygen model proposes that carefully fabricated $Am_2CuO_4$ doped with Ce (or possibly with Th) will superconduct with $T_c \approx 24\,K$, provided the sample geometry permits one to overlook the sample heating by the radioactivity of Am. (Use of freshly synthesized $Am_{2-z}Ce_zCuO_4$ with $^{243}Am$ instead of $^{241}Am$ will limit radiation damage and make the cooling problems of radioactive Am about a factor of 18 less severe: 6.3 mW/g.[13])

## II. CHARGE-RESERVOIR OXYGEN MODEL

The essential elements of the charge-reservoir model are.[9] (i) The primary superconducting condensate lies in the charge-reservoir layers of the various crystal structures (i.e., in the Am-O layers of $Am_{2-z}Ce_zCuO_4$), not in the cuprate planes. (ii) In the $T'$-structure $R_{2-z}Ce_zCuO_4$ homologues, interstitial oxygen is a prerequisite of superconductivity and dopes the host material *p* type.[8] (iii) The physics of superconductivity is nearly Bardeen-Cooper-Schrieffer-like (BCS-like),[14] but with polarization pairing by bosons that are nonmagnetic: phonons, polarons, excitons, or plasmons (as opposed to spin-fluctuation pairing, for example[15]). (iv) The Cooper pairs are broken by Abrikosov-Gor'kov magnetic scattering.[16] Hence the model contrasts with all cuprate-plane models in that (i) it locates the main superconducting condensate in the charge-reservoir layers and (ii) it also *requires* the presence of interstitial oxygen in the $R_{2-z}Ce_zCuO_4$ materials, which causes these materials to conduct *p* type.[8] It also disagrees with those many cuprate-plane models that hypothesize either magnetic pairing or major deviations from Abrikosov-Gor'kov pair breaking.

### A. Crystal-field splitting in Th- or Ce-doped $R_2CuO_4$

In the charge-reservoir picture, the failure of $Cm_{2-z}Th_zCuO_4$ to superconduct is attributed to the $L=0$ nature of $Cm^{3+}$ in $Cm_{2-z}Th_zCuO_4$, and the resulting occurrence of pair breaking that is uninhibited by crystal-field splitting (the crystal-field splitting inhibits pair breaking by $L \neq 0$ rare-earth and actinide ions, but there is no splitting by $L=0$, $J \neq 0$ magnetic ions such as $Cm^{3+}$—where $L$ and $J$ are total orbital and overall angular momentum quantum numbers[17–19]). Experiments supporting this viewpoint include the fact that (i) the isoelectronic compound

$Gd_{2-z}Ce_zCuO_4$ does not superconduct either, because $Gd^{3+}$, like $Cm^{3+}$, has $L=0$ and $J\neq 0$; (ii) the alloys $R_{2-z}Ce_zCuO_4$ do not superconduct for $R=La_{1-x}Gd_x$,[20–22] $R=Nd_{1-x}Gd_x$,[21,23,24] or $R=Eu_{1-x}Gd_x$ (Refs. 24 and 25) (because the $L=0$ Gd pair breaker is present in significant amounts), but (iii) the alloys $R_{2-z}Ce_zCuO_4$ *do superconduct* for $R=La_{1-x}Nd_x$ or for $R=La_{1-x}Eu_x$ (Refs. 20, 21 and 26–28) because pair-breaking $L=0$ rare-earth ions such as $Gd^{3+}$ are absent.[24,29,30] [These facts *cannot* be organized in terms of the average radius of the trivalent ion $R$,[24] because some nonsuperconducting compounds with $R=La_{1-x}Gd_x$ (Refs. 20–22) have even larger average rare-earth radii than Nd of superconducting $Nd_{2-z}Ce_zCuO_4$.]

The charge-reservoir picture postulates that the superconducting condensates of the $T'$-structure $R_{2-z}Ce_zCuO_4$ homologues involve interstitial oxygen sandwiched between the $O_2$ layer in a unit cell and a cuprate plane. This oxygen dopes the host $p$ type.[8]

### B. Pair breaking by $Cm_{Ba}$ in $CmBa_2Cu_3O_7$

The charge-reservoir model has $CmBa_2Cu_3O_7$ superconducting when pure, but not when normally prepared, because it contains too many $Cm_{Ba}$ defects which are rather more soluble on Ba sites than $Gd_{Ba}$ defects in $GdBa_2Cu_3O_7$, for example.[6,31] As with Pr in $PrBa_2Cu_3O_7$,[2] the large magnetic ions of Cm are highly soluble on the Ba sites, where they break Cooper pairs and destroy the superconductivity: only if the number of Ba-site rare-earth defects $Pr_{Ba}$ or $Cm_{Ba}$ is minimized will $PrBa_2Cu_3O_7$ or $CmBa_2Cu_3O_7$ superconduct. Pure $PrBa_2Cu_3O_7$ superconducts,[2,3,32–43] while the same material with ~10% or more $Pr_{Ba}$ defects does not. Signatures of the $Pr_{Ba}$ defects include (i) a Néel temperature of 17 K [no Néel transition has been detected in the superconducting material down to 4 K (Refs. 44 and 45)], (ii) significant occupancy of the O(5) antichain oxygen site, and (iii) excess $BaCuO_2$ during sample fabrication. $CmBa_2Cu_3O_7$ also has a high Néel temperature $T_N\approx 22\,K$,[46] indicative of significant numbers of the pair-breaking $Cm_{Ba}$ defects.[47]

The situation is similar in $Pr_{2-z}Ce_zSr_2Cu_2NbO_{10}$,[13,48–54] which has a similar crystal structure and has significant concentrations of pair-breaking $Pr_{Sr}$ defects—but has recently exhibited (granular) superconductivity:[6,55] this material's former lack of superconductivity is now attributable to the large size of the rare-earth ion $Pr^{3+}$, its high solubility on the Sr site (recently measured[55]), and pair breaking.

### C. Superconductivity in $Am_{2-z}Ce_zCuO_4$

$Am_{2-z}Ce_zCuO_4$ should superconduct, barring unforeseen difficulties associated with the radioactivity of Am.

Moreover, $Am_{2-z}Ce_zCuO_4$ homologues obtained by replacing $Am^{3+}$ with a trivalent rare-earth ion will superconduct, unless (i) a size effect associated with the rare-earth (or Am) ion prevents the host from forming a cage large enough to enclose interstitial $O^-$ between the $O_2$ layer and the cuprate plane[56] or unless (ii) crystal-field splitting is impossible for the *magnetic* rare-earth ion replacing $Am^{3+}$, because it has $L=0$ (such as for $Gd^{3+}$ or $Cm^{3+}$), and so pair breaking is effective.[18,19]

The $L=0$ pair-breaking effect due to the absence of crystal-field splitting must occur for *both* the Gd and Cm homologue; it cannot explain why the Gd homologue superconducts (e.g., $GdBa_2Cu_3O_7$), while the Cm homologue ($CmBa_2Cu_3O_7$) does not. In those classes of materials whose Gd homologue does superconduct, e.g., $GdBa_2Cu_3O_7$ or $Gd_{2-z}Ce_zSr_2Cu_2NbO_{10}$, the superconducting condensate must lie more than one nearest-neighbor distance from the Gd ions in order to provide superconductivity unbroken by the Gd magnetic moment (in the vicinity of the Cu-O chains and the Ba-O layers of $GdBa_2Cu_3O_7$ or near the Sr-O layers of $Gd_{2-z}Ce_zSr_2Cu_2NbO_{10}$). With the crystal-field splitting effect ruled out for $CmBa_2Cu_3O_7$ by the superconductivity of $GdBa_2Cu_3O_7$, we proposed $Cm_{Ba}$ pair breaking as the mechanism for the failure of $CmBa_2Cu_3O_7$ to superconduct.[18,19]

### D. Charge states of Am and Ce

The next question is, does Am in $Am_{2-z}Ce_zCuO_4$ assume the $Am^{3+}$ valence state? This question is important, because some compounds, such as $Pb_2Sr_2AmCu_3O_8$,[57] do not superconduct on account of Am being $Am^{4+}$ rather than $Am^{3+}$ (''hole filling'').

The ionization potential of Am from the $Am^{3+}$ to the $Am^{4+}$ state is known[58] to lie between those of Ce (36.76 V) and Pr (38.98 V),[59] and the magnitude of the Madelung potential at the rare-earth site is approximately 29.85 V.[6] Consequently, Am will be in the $Am^{3+}$ charge state in $Am_2CuO_4$.

The nature of Ce doping is different in the charge-reservoir and cuprate-plane models. In the charge-reservoir model, Ce bonds with an adjacent *interstitial* oxygen and forms $Ce^{4+}$ paired with the interstitial: a $(Ce, O_{interstitial})$ pair that is a net $p$-type dopant.[8,56,60] Without the interstitial oxygen, the magnitude of the Madelung potential at the Ce site is too weak by ~7.7 V to ionize $Ce^{3+}\rightarrow Ce^{4+}$. With the interstitial oxygen, the Madelung potential is just barely strong enough to ionize Ce to $Ce^{4+}$. (In the cuprate-plane models, Ce is *assumed* to be in the $Ce^{4+}$ charge state, but no reason for this ionization is given, and so these models are not self-consistent.)

Since the magnitude of the ionization potential of Am is larger than that of Ce and since Ce is just barely ionized to $Ce^{4+}$ by interstitial oxygen, we believe that Am cannot self-dope $Am_2CuO_4$ by attracting interstitial oxygen and forming $(Am^{4+}, O_{interstitial})$ pairs. But a prudent investigator should perform measurements of the magnetism of $Nd_{2-z}Am_zCuO_4$ to guarantee that Am itself does not become a dopant.[61]

### E. Hybridization

Many authors appealed to hybridization of the Pr $4f$ and O $2p$ orbitals as a way of explaining why $PrBa_2Cu_3O_7$ did *not* superconduct,[62] and so it would be remarkable if hybridization were the reason that $Am_{2-z}Ce_zCuO_4$ superconducts. Zou *et al.*[39] and Ye *et al.*[39] have convincingly shown that hybridization does not quench the $PrBa_2Cu_3O_7$ superconductivity: Both by fabricating single crystals that superconducted and by measuring the pressure dependence of $T_c$ (which *increased* with pressure, contrary to the expectations of hybridization theory[34,39,61]), they demonstrated that hybridization is neither operative nor important. To our knowledge, no convincing evidence exists of any degradation of $T_c$ by hybridization in $4f$ or $5f$ high-temperature superconductors.[62] Furthermore, Soderholm *et al.*[30] have argued that the $5f$ electrons of Cm are more localized than the $4f$ electrons of Pr, which means that the failure of $CmBa_2Cu_3O_7$ to superconduct cannot be ascribed to hybridization, since the failure of $PrBa_2Cu_3O_7$ to superconduct was *not* due to hybridization.

If hybridization of the actinide $5f$ wave function were to be important (and it could conceivably be important, because the actinide $5f$ radii are larger than their $4f$ counterparts), one would expect hybridization to *quench* superconductivity in $Cm_{2-z}Th_zCuO_4$, for the same reasons that hybridization purportedly quenched superconductivity in $PrBa_2Cu_3O_7$ (before clean material was fabricated). One way to examine this issue is to prepare $(Cm_{1-x}Am_x)_{2-z}Th_zCuO_4$, which should superconduct with increasing $x$. However, this experiment would only discriminate between cuprate-plane and charge-reservoir mechanisms by the functional form of $T_c(x)$, because less pair breaking in a charge-reservoir model and less hybridization in a cuprate-plane model should both increase $T_c$ with $x$. The more discriminating test is with measurements of interstitial oxygen.

### F. Interstitial oxygen

Distinguishing features of the charge-reservoir model of superconductivity are that (i) the carriers of superconductivity are holes (which contribute to *positive* Hall and thermopower coefficients,[63] although only the most perfect crystals exhibit these properties) and (ii) the crystal must contain at least several percent interstitial oxygen per unit cell.

Most cuprate-plane theories of $Nd_{2-z}Ce_zCuO_4$ homologues regard these homologues as doped $n$ type (without explaining how this can be), but in the charge-reservoir model, the doping is necessarily $p$ type and requires interstitial oxygen as a condition of superconductivity. It should be possible to discriminate between these two models by removing interstitial oxygen. Superconducting $Am_{2-z}Ce_zCuO_4$ should continue to superconduct in a cuprate-plane picture, but will lose its superconductivity in a charge-reservoir model.

## III. CUPRATE-PLANE MODEL PREDICTIONS

We are unaware of any explicit predictions based on any cuprate-plane model of superconductivity for the expected behavior of $Am_{2-z}Ce_zCuO_4$. But we do note that any such predictions must *simultaneously* explain or predict, (i) whether $Am_{2-z}Ce_zCuO_4$ will superconduct; (ii) why $Cm_{2-z}Th_zCuO_4$ does not superconduct;[30] (iii) why $Gd_{2-z}Ce_zCuO_4$ does not superconduct;[8] (iv) why $CmBa_2Cu_3O_7$ does not superconduct;[46] (v) why $GdBa_2Cu_3O_7$,[64] $Gd_{2-z}Ce_zSr_2Cu_2NbO_{10}$,[55] and $Pb_2Sr_2Gd_{1-x}Ca_xCu_3O_8$ (Ref. 65) do superconduct; and (vi) how Ce can dope any $R_2CuO_4$ compound either $p$ or $n$ type.

The Ce-doping issue is especially interesting: Ce doping is *assumed* to produce both $n$-type doping and $Ce^{4+}$ in isolation [although (i) no demonstration has been presented showing that the Madelung potential at a Ce site can be consistent with a $Ce^{4+}$ charge state and (ii) no explanation has been given of why $YBa_2Cu_3O_7/Nd_{2-z}Ce_zCuO_4$ junctions do not rectify, despite being purportedly $p$-$n$ junctions[66]].

All cuprate-plane theories place the primary superconducting condensate in the cuprate planes. Many, but not all, assume spin-fluctuation pairing (already ruled out experimentally[15]) or that either Abrikosov-Gor'kov pair breaking or crystal-field splitting are inoperative or unimportant.

In our opinion, most cuprate-plane theories attempting to describe the facts about superconductivity for Cm-, Am-, and Gd-based compounds will conclude that $Am_{2-z}Ce_zCuO_4$ *will not* superconduct. There is a clear difference between the predictions of such cuprate-plane theories and the charge-reservoir oxygen model. (This should be the case for most of those cuprate-plane theories that accept the nonsuperconductivity of $Cm_{2-z}Th_zCuO_4$ or of $CmBa_2Cu_3O_7$.)

In the less likely alternative that a specific cuprate-plane theory claims $Am_{2-z}Ce_zCuO_4$ *must superconduct* (or, once it does superconduct, if some cuprate-plane theory claims to explain why), then attention should be drawn to whether this theory is also capable of explaining why $Cm_{2-z}Th_zCuO_4$,[30] $Gd_{2-z}Ce_zCuO_4$,[67] and $CmBa_2Cu_3O_7$ (Ref. 46) do not superconduct, while $GdBa_2Cu_3O_7$,[64] $Gd_{2-z}Ce_zSr_2Cu_2NbO_{10}$,[55] and $Pb_2Sr_2Gd_{1-x}Ca_xCu_3O_8$ (Ref. 65) do superconduct. Any attempt to explain the nonsuperconductivity of $Cm_{2-z}Th_zCuO_4$ (or $Gd_{2-z}Ce_zCuO_4$) in terms of crystal-field splitting would also have to explain the superconductivity of the other Gd compounds: $GdBa_2Cu_3O_7$, $Gd_{2-z}Ce_zSr_2Cu_2NbO_{10}$, and $Pb_2Sr_2Gd_{1-x}Ca_xCu_3O_8$. Why do the almost identical cuprate planes, which experience virtually the same bonding to Gd in all of these compounds, produce some materials which superconduct and others which do not?

The explanation that $Cm_{Ba}$ destroys the superconductivity in $CmBa_2Cu_3O_7$, while $Cm_{Cm}$ does not, is not available to a cuprate-plane model for the same reason that it is unavailable for $PrBa_2Cu_3O_7$:[2] the primary superconducting condensate cannot occupy the cuprate planes that are essentially midway between the Cm and Ba-O layers and yet have $Cm_{Ba}$ and $Cm_{Cm}$ scatter Cooper pairs differently.

Hence, even if $Am_{2-z}Ce_zCuO_4$ superconducts, cuprate-plane models will still be faced with answering the question, why do $Cm_{2-z}Ce_zCuO_4$ and $CmBa_2Cu_3O_7$ not superconduct?

## IV. CONCLUSION

Regardless of which set of (consistent) predictions are made for the cuprate-plane picture, in the charge-reservoir oxygen model of high-temperature superconductivity, $Am_{2-z}Ce_zCuO_4$ is predicted to superconduct if it is doped such that we have $z \approx 0.15$—barring unforeseen problems with radioactivity. A demonstration of such superconductivity would lend further support to this model and would place an added burden on advocates of cuprate-plane theories to explain the lack of superconductivity in $Cm_{2-z}Th_zCuO_4$,[30] $Gd_{2-z}Ce_zCuO_4$,[67] and $CmBa_2Cu_3O_7$,[46] as well as the superconductivity of $GdBa_2Cu_3O_7$, $Gd_{1-z}Ce_zSr_2Cu_2NbO_{10}$, and $Pb_2Sr_2Gd_{1-x}Ca_xCu_3O_8$.

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