these minerals serve as convenient neutron dosimeters for the present purposes.

In short, uniform, well-located glass is the material of choice. I would welcome communication from anyone who knows of such material, or who can direct me to someone who might, at the Department of Earth and Environmental Sciences, Rensselaer Polytechnic Institute, 110 8th Street, West Hall G-17, Troy NY 12180-3590; phone 518-276-8523; fax 518-276-8627.

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10/93

In Explaining High T_c, Is d-Wave a Washout?

The Search and Discovery story "In High-T_c Superconductors, Is d-Wave the New Wave?" (May 1993, page 17) did not adequately reflect the nature and severity of the negative comments that I expressed to its author, Barbara Goss Levi. I believe my misgivings are widely shared in the theoretical community and I think they should have been less casually treated.

Levi's treatment of the experiments, which occupied most of the article, was careful and pretty much evenhanded. One not entirely minor point is that the photoemission spectrum of Zhi-xun Shen given in the striking illustration on page 19 is also a striking illustration of how lines drawn to "guide the eye" often deceive it instead. The blue and green points (obtained above and below T_c, respectively) on curve B differ by more than experimental error and clearly signal an energy gap in the Fermi surface of some magnitude at point B in wavevector space. (I estimate approximately 0.3 of the gap at point A.) The line as drawn is not a good representation of either set of points. Thus while the evidence for anisotropic electron pairing is strong, that for a node in the gap, as expected for $d_{x^2-y^2}$ pairing, is not.

I also feel that the remark quoted from Malcolm Beasley seriously misstates the situation: Many of us feel that experiments have *already* told us a great deal.

While the theoretical section of the story is brief, the prominence given the experiments is clearly motivated by theory, so the following points should be taken into consideration:

▷ The "spin fluctuation" theories of T. Moriya, Douglas Scalapino and David Pines, especially that of Pines, are in a real sense not true theories but rather heuristic models with many unexamined assumptions. Some of those assumptions are very questionable, particularly the assumption that antiferromagnetic spin fluctuations can result from a perturbative "Fermi liquid" model. Antiferromagnetism as normally observed is a consequence of the Mott–Hubbard gap, which cannot be treated perturbatively. (The exchange coupling J is proportional to $1/U$, which cannot arise perturbatively in U, the interaction coupling constant.) Another, related assumption is the neglect of vertex corrections, as pointed out by J. Robert Schrieffer in his talk at the Santa Fe meeting where the problem of spin-fluctuation theory was extensively discussed.

▷ The spin-fluctation model relies heavily on detailed computer calculations that are not subject to independent check and that have unknown sensitivity to the choice of parameters. Not only are a considerable array of arbitrary parameters adjusted to fit the data, but the spin fluctuations enter into the calculations via an arbitrary function that is only weakly determined by experiment. These computer fits are carried out for normal-state transport and magnetic properties as well as superconducting properties, and their workings are not available for detailed examination. In contrast, the most seriously competing theory, my own, relies on no calculations that cannot be verified by anyone on the proverbial back of an envelope (except for the recent calculations of gap anisotropy by Sudip Chakravarty, Asle Sudbø, Steven Strong and me). The few parameters used are not sensitive and are mostly commonsensical. Even the gap anisotropy (which was not produced in response to the experiments, contrary to Levi's statement) follows from a few-parameter tight-binding model that fits the calculated band structure. Normal-state properties are all simple power laws with the power determined from first principles. The fact that my approach is theoretically deep leads to computational simplicity.

▷ The normal states of all of the high-T_c materials at optimal doping differ very little; for instance, as shown by Bertram Batlogg, the resistivity per plane at optimal doping is almost independent of T_c as T_c varies from approximately 5 K to approximately 125 K. Many other normal-state properties are simple, anomalous and generic. This makes it implausible that the same interactions that cause T_c are responsible for the normal-state resistivity (for instance) in all cases, as postulated by the spin-fluctuation theories. T_c seems to have no correlation with magnetic fluctuations or with other normal-state properties. The properties of high-T_c materials are, heuristically, very simple and very striking. For instance, I have shown that a single, simple formula based on interlayer interactions can parametrize the T_c's of all known materials. (It fits the new mercury-based materials very well.) The use of laborious case-by-case calculations with no clear heuristic guidelines seems misguided in the face of this simple phenomenology. The conventional attitude, that laborious calculations lend weight to a theory, is opposite to the reality: that valid theories are invariably subject to many simple, qualitative checks.

▷ It seems impossible that superconductivity is caused by spin fluctuations, in view of the sensitive dependence of T_c on the nonmagnetic constituents of the material. Nonetheless, as I have pointed out, the symmetry of the gap function depends on residual interactions and could (though I think it unlikely) be d-wave. I commented to Levi on at least two contrary indications: the impurity insensitivity of the copper oxides and the isotope effect seen in lower-T_c examples such as $(La\text{–}Sr)_2CuO_4$, which has normal sign, if reduced magnitude compared with ordinary elemental superconductors. Levi quoted only the former. The isotope effect would be likely to be of anomalous sign for a d-wave gap, since phonon scattering is pair-breaking for such a gap. Pines and Philippe Monthoux cannot calculate the impurity effect as they claim, since their theory has no explanation for the absence of residual resistance in, for instance, this substance.

The spin-fluctuation "theory" has been welcomed by experimentalists because it appears simple conceptually and incorporates many experimental data. Some of the data are sound—the nmr data, for instance—but not uniquely interpretable; the neutron data are straightforward in principle but very sketchy in detail: For instance, no magnetic signal identified with the "chain" electrons is ever seen. There are enormous backgrounds, and samples of the appropriate size are seldom unequivocally pure. The theory ignores, among other data, the extensive and striking photoemission data on normal-state samples, which seem incompatible with simple perturbative "Fermi liquid" theories.

It is hard to understand why such a facile but naive approach should be taken so seriously, even if, hopefully, only briefly so. I do not attempt to explain this phenomenon, but it is not scientific in origin. There is no possibility that spin-fluctuation theory in the current form can explain high-T_c cuprates.

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6/93

PINES REPLIES: The physical origin and theoretical description of high-temperature superconductivity will be settled in the refereed scientific literature, not in the Letters department of PHYSICS TODAY. However, it is important to respond to Philip Anderson's letter, since he has made such a strong attack on the work of so many people whose scientific credentials I hold in esteem and on a general theoretical viewpoint, the spin-fluctuation mechanism with its predicted $d_{x^2-y^2}$ pairing state, that is backed by a rapidly growing body of theoretical and experimental work published in that refereed literature.

In summary, and in direct contradiction to Anderson's assertions, experiment demonstrates that antiferromagnetic spin fluctuations play a key role in determining normal-state behavior, which does not consist of simple power laws, and makes it evident that computer calculations are essential if one seeks a quantitative account of experiment. For a model Hamiltonian of planar quasiparticles coupled by the anomalous spin fluctuations that their magnetic interaction produces, the present generation of computer calculations provides a surprisingly accurate account of the resistivity, optical properties and superconducting transition temperatures T_c of $YBa_2Cu_3O_7$, and of the changes in the resistivity and in T_c when Ni and Zn impurities substitute for the planar Cu sites in this system.

As Barbara Goss Levi noted in her Search and Discovery story, many theoretical groups have explored ways of using spin fluctuations as a source of high-temperature superconductivity. Our theoretical research in Urbana,[1] carried out in collaboration with Alexander Balatsky, Victor Barzykin, Andy Millis, Hartmut Monien, Philippe Monthoux, Alexander Sokol and Dean Thelen, is based on the premise that the spin and charge properties of $YBa_2Cu_3O_7$ and other cuprate superconductors reflect the close approach of the normal state to anitferromagnetism. Strong support of this *Ansatz* has been provided by experiments on $YBa_2Cu_3O_{6.63}$[2] and $YBa_2Cu_4O_8$[3] that show that the spin-gap-induced changes in the spin-fluctuation spectrum measured in nmr experiments are accompanied by changes in the normal-state resistivity and Hall coefficient, and by recent nmr experiments[4] and accompanying theoretical work[5] that demonstrate that in the normal state the anomalous magnetic properties of the cuprate superconductors reflect different (as a function of doping and as a function of temperature) regimes of scaling behavior, similar to those found in calculations of the influence of holes on the two-dimensional Heisenberg antiferromagnetic description of the insulating state.[6] The physical origin of this scaling behavior is the near-antiferromagnetism of the normal state.

Realistic calculations on the cuprate superconductors must therefore take into account nonperturbatively both the nonlinear feedback effects responsible for the measured magnetic scaling in the metallic state and the structure in momentum space of the spin-fluctuation-induced interaction between quasiparticles, which, if neglected, leads one to underestimate T_c by factors of three to five.[1] Such calculations cannot be carried out analytically; one has no choice but to rely on the computer. To study these effects in $YBa_2Cu_3O_7$, Monthoux and I have used a model Hamiltonian in which the quasiparticle and spin excitation spectra are taken from experiment. In work that is easily subject to independent verification by a computationally literate theorist, we found that by using fast-Fourier-transform techniques, we could obtain accurate solutions of the coupled nonlinear Eliashberg equations on a Cray Y-MP.[1] In our most recent work,[7] we explore the sensitivity of T_c to the details of the spin spectrum, calculate impurity-induced changes in the resistivity and T_c, and explain the remarkable difference in the influence of Ni and Zn impurities on T_c. That difference serves as a "smoking gun" for the spin-fluctuation mechanism, since Zn, which changes the local magnetic order and hence the spin-fluctuation-induced interaction responsible for superconductivity, has a three times greater influence on T_c than does Ni, which scarcely affects the low-frequency magnetic properties.[8]

As Monthoux, Balatsky and I have frequently emphasized, since our nearly antiferromagnetic Fermi-liquid approach predicts unambiguously that the superconducting state of $YBa_2Cu_3O_7$ (and any other system for which the exchange of spin fluctuations peaked at the commensurate antiferromagnetic wavevector provides the superconducting mechanism) *must* be $d_{x^2-y^2}$, experimental detection of that pairing state is a crucial test of our theory. Two recent additions to the experiments[9,10] discussed by Levi that support d-wave pairing are the work of Thomas Deveraux and colleagues,[11] who find that measurements of the electronic Raman effect provide a large amount of symmetry-dependent information, with results for $Bi_2Sr_2CaCu_2O_8$ in agreement with $d_{x^2-y^2}$ pairing, and of Dermot Coffey,[12] who has reviewed the considerable body of evidence in tunneling experiments for features, predicted by $d_{x^2-y^2}$ pairing, at multiples of the superconducting energy gap.

The extreme sensitivity to sample purity found in measurements of the low-temperature properties of the superconducting cuprates also finds a natural explanation in $d_{x^2-y^2}$ pairing, since at low temperatures imperfections change the clean-limit density of states for quasiparticles near the point nodes of the two-dimensional system to a constant value.[13] The sample sensitivity revealed in controlled nmr[8] and penetration depth experiments[9] explains why so many previous experimentalists working with thin films or somewhat imperfect crystals have found a quadratic variation for penetration depth at low temperatures.

Of course much more theoretical and experimental work remains to be done before we arrive at a full understanding of superconductivity in the cuprates. We are, however, at a stage where the quality of the experimental work demands more than back-of-the-envelope calculations by theorists. Indeed, "the devil is in the details," which show, for example, that the magnitude of the gap contained in the anisotropic s-wave state of Sudip Chakravarty and colleagues[14] is too large to be *quantitatively* compatible with nmr and penetration depth experiments on $YBa_2Cu_3O_7$.[9] It is to be expected that the spin-fluctuation mechanism and $d_{x^2-y^2}$ pairing will continue to present a tempting target for experimentalists and theorists alike, and those of us who believe d-wave is not only the new wave but the right wave can only welcome that attention.

continued on page 120

continued from page 15

In preparing this response I have relied heavily on the written version of a review talk presented at the La Jolla Conference on Strongly Correlated Electron Systems,[15] to which the interested reader is referred for further details.

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1/94

SCALAPINO REPLIES: I believe that there are sound theoretical and experimental reasons for considering the possibility that the pairing mechanism in the cuprate superconductors is associated with antiferromagnetic spin fluctuation that leads to a $d_{x^2-y^2}$ gap.

While the initial theoretical suggestions for this mechanism were based upon perturbation theory for a two-dimensional Hubbard model doped near half-filling, subsequent Monte Carlo calculations[1] have shown that there is an attractive pairing interaction in the $d_{x^2-y^2}$ channel. Furthermore, exact diagonalization studies[2] of the strong-coupling t–J limit of the Hubbard model provide evidence that two holes form a $d_{x^2-y^2}$ bound state when the ratio of the exchange coupling J to the hopping t is greater than a critical value. Although none of these numerical calculations has provided definitive evidence for a $d_{x^2-y^2}$ superconducting state, the fact that quite different numerical approaches, ranging from conserving diagrammatic approximations to Monte Carlo and Lanczos numerical studies, all find evidence for $d_{x^2-y^2}$ pairing correlations is significant.

With respect to experiment, a variety of results have been compared with the spin-fluctuation $d_{x^2-y^2}$ pairing ideas, because detailed, albeit approximate, calculations based on those ideas have been carried out. Again, these phenomenological calculations certainly don't provide a unique interpretation, but the range of phenomena that have been fit within this framework is striking. For example, in addition to the nmr longitudinal-relaxation-time measurements mentioned in the Search and Discovery story, the temperature dependence of the transverse relaxation time[3] supports a $d_{x^2-y^2}$ scenario. In addition, the same form for the spin susceptibility that was used in calculating these nmr relaxation times provides a $d_{x^2-y^2}$-based explanation[4] for the isotropic but incomplete suppression of the neutron scattering intensity experimentally observed[5] in $La_{1.86}Sr_{0.14}CuO_4$ below T_c.

Recently an analysis of the temperature dependence of the microwave penetration depth and its dependence on impurities[6] has shown that a $d_{x^2-y^2}$ gap also provides a plausible explanation for why the temperature dependence of the penetration depth observed at low temperatures in clean YBCO is linear, while a quadratic dependence appears when impurities are added. A similar analysis that has been carried out for the real part of the microwave conductivity[7] will test whether the dynamic and impurity scattering lifetime effects are consistent with the $d_{x^2-y^2}$ and spin-fluctuation ideas.

Thus the question of whether the cuprate superconductors are in a $d_{x^2-y^2}$ pairing state induced by the underlying short-range antiferromagnetic correlations will be decided experimentally.

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10/93

SHEN REPLIES: The figure to which Philip W. Anderson refers was poorly reproduced in PHYSICS TODAY. Readers should refer to our original figure and discussion[1] for experimental details. We discussed the gap anisotropy in the context of the position of the midpoint of the leading edge of the photoemission intensity peak. At point A, the midpoint of the leading edge is shifted to higher binding energy below T_c, indicating a gap opening. At B, the midpoint is not shifted within the uncertainty, reflecting a much smaller (or null) gap. We need more theoretical input to understand the change of the photoemission line shape as a function of temperature.

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A Record-Breaking Superconductor, Missed

I read with interest the news story [by Barbara Goss Levi] in the July 1993 issue (page 20) entitled "Critical Temperature Nears 135 K in a Mercury-Based Superconductor." I was surprised by the opening sentences: "No superconductor has broken the record for the highest critical temperature since 1988, when a thallium-bearing compound, exhibiting resistanceless conduction at tempera-