High T_c and the Materials Genome

i) High-Pressure Hydrogen Sulfide from First Principles: A Strongly Anharmonic Phonon-Mediated Superconductor
Ion Errea, Matteo Calandra, Chris J. Pickard, Joseph Nelson, Richard J. Needs, Yinwei Li, Hanyu Liu, Yunwei Zhang, Yanming Ma and Francesco Mauri, Phys. Rev. Lett. **114**, 157004 (2015)

ii) Perspective: Role of Structure Prediction in Materials Discovery and Design
Richard J. Needs and Chris J. Pickard, APL Materials **4**, 053210 (2016).

Recommended with a commentary by Zachary Fisk (U. C. Irvine), Warren E. Pickett (U. C. Davis) and Joe. D. Thompson (Los Alamos)

Not only was the discovery of MgB_2 a surprise, so also was the speed with which theory was able to rationalize its remarkable T_c. The more recent spectacular discovery [1] of superconductivity in high pressure H_2S has further highlighted the remarkable ability of theory to both identify H_3S as the superconducting phase [2] and now, in the paper of Errea *et al*. [3], the importance of anharmonicity in limiting the electron-phonon λ. The search in hydrogen-based materials certainly owes much to vision of Neil Ashcroft [4], and there are now suggestions that superconductivity has been possibly observed at room temperature in hydrogen at 500 GPa.[5] Clearly modern structure theory has shown its power here, a power that was not available until fairly recently. Such theoretical prowess might be supposed to provide the path to new, higher temperature superconductors but the *a priori* success in finding new superconductors via such calculations has been virtually nil.

But nobody knows where to find new superconductors and so far theory even in its new powerful versions has been no real help. With the above materials we are dealing with BCS superconductors, and here the good news is there is no need to invent a paring mechanism that may or may not be relevant. From the standpoint of materials, superconductivity is a chemistry problem, not one of physics. As was suggested in the failed Frohlich theory of superconductivity, higher T_c and lattice instability go hand in hand, so one way to think about BCS superconductivity is that it is a diagnostic of an incipient lattice stability problem. However, even in the area of superconducting materials, the success of theory in discovering new superconductors is really quite modest, and this raises the question as to how the computing capabilities now apparent might be put to more effective use. Consider the following example. Johnson and Jeitschko (1974) [6], through what can be described as classic solid state chemical thinking, designed and synthesized the filled PbFCl variant ZrCuSiAs. Subsequently, Jeitschko's student Zimmer in her thesis work (1995) produced LaFeOP in this structure and found, as reported in her thesis but not in the publication resulting from it [7], superconductivity near 5 K. The F-doping of this material by Hosono and collaborators (2008) [8] is what sparked the pnictide superconductivity iron rush. Why were the superconducting possibilities in this class of materials not apparent to Johnson and Jeitschko when they first synthesized the compound prototype? It must be that the ghost

of superconductivity is already present there and surely a sufficiently effective computation should reveal this. Richard Hamming's observation seems to the point here: "The purpose of computation is insight, not numbers." [9] From a bench scientist's perspective the really useful development from theory will be the honing of one's materials instinct. The work of Needs and Pickard [10] show the power computation now has to predict new structures. But this impressive success is not accompanied by deeper insight. Superconductivity from the materials standpoint might be thought of as a manifestation of an internal "tension" in a material, a "tension" that is not otherwise so obvious and that we could hope might be revealed through a clever computational approach (yet to be envisioned?). An interesting paper in the 1970's by Cohen and Anderson on maximum T_c noted that very large λ started to look like a covalent bond.[11] Does this lie hidden within the simple low T_c pnictides?

With phonon-driven T_c's now exceeding those of materials believed to have T_c arising from non-phonon mechanisms, it makes sense to push further the frontiers of the conventional superconducting materials. What the bench scientist needs is a deeper materials savvy to guide the search for the other MgB_2's that might be out there. This seems the place where these superior computational tools now in place can educate us about superconductors in an entirely new way.

References

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