CORRELATIONS BETWEEN BOND STRUCTURE AND THERMODYNAMICS IN ORGANIC SUPERCONDUCTORS

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We have recently proposed a mechanism of superconductivity (SC) in strongly-correlated ¼-filled materials based on frustration-induced mobility of real-space singlet pairs [1,2]. A critical parameter in theories where SC follows the Schafroth proposal [3] of charged bosons is the effective mass of the singlet boson pairs. For weak singlet pair binding the superconducting critical temperature T_c is too low; but for binding that is too strong, pair mobility is low and the material is a spin-gapped insulator.

In this talk we focus on organic charge transfer solids (CTS). The common feature of superconducting CTS is ¼-filled band. However, not all ¼-filled band CTS are superconducting and there is currently no understanding of what distinguishes the superconductors from the non-superconducting semiconductors. To understand this difference we have examined the the metal-insulator (MI) transitions in CTS that lead antiferromagnetism, charge order or spin gap (SG). We note that there are two paths to the semiconducting ground state in the CTS: (i) via a single thermodynamic transition where the MI and SG transitions occur simultaneously, and (ii) via separate transitions where a high-temperature MI transition is followed by a low-temperature SG transition. In one dimension (1D), the occurrence of one versus two transitions depends on the strength of Coulomb interactions. Beyond 1D we show that in a quasi-1D ladder a single combined transition occurs regardless of Coulomb interaction strength [4]. Although the charge and bond strengths exhibit periodicity 4 in all cases, the phase relationships between them are different. Thus in all cases pairs of charge-rich sites are followed by pairs of charge-poor sites, but the bond strength is largest between the charge-rich sites for (i) and between a charge-rich and a charge-poor site in (ii).

We survey a large number of ¼-filled CTS and related inorganic materials with spin-gapped and superconducting states and find an empirical relationship: in materials with the strongest bonding between the charge-rich sites (so-called "intra-dimer" singlet formation) SG transitions tend to occur at high temperature and simultaneously with the MI transition [4]. These materials tend to remain insulating rather than becoming superconductors under the application of pressure. Conversely, materials with weaker singlet binding ("inter-dimer" singlet formation) show thermodynamically distinct SG and MI transitions and tend to become superconductors under the application of pressure. This phenomenology is difficult to explain within mean-field spin or charge-fluctuation theories of SC and lends further support to our proposed mechanism [1,2].

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