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Exploring high-temperature superconductivity in hard matter close to structural instability

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ABSTRACT

This paper discusses where and how high-temperature superconductors can be found.

I. INTRODUCTION

Ever since the discovery of superconductivity in mercury by Heike Kamerlingh Onnes in 1911, it has been hoped that a general rule can be found to guide the search for new superconductors with higher critical temperature T_c. Matthias's rules have turned out to be very successful in finding superconductivity in transition metal compounds but have not worked for many other materials. This paper is devoted to the exploration of superconductivity with a high T_c, even above room temperature, in materials near lattice instability on the basis of a soft mode picture in which the soft modes involved in the instability are assumed to be particularly effective in enhancing T_c. The concept of a "soft mode" provides a basis for understanding the dynamics of the processes involved in a phase transition. For phase transitions of second or first order close to the tricritical point, the frequency of each mode of a crystal lattice tends to zero or to substantially decrease.[1]

II. WHAT WE HAVE LEARNED FROM OLD, NEW, AND THE NEWEST HIGH-T_C SUPERCONDUCTORS

Before the discovery of superconductivity in cuprates, the A15 systems were considered as high-T_c superconductors. These systems possess many anomalies in both their superconducting and normal states compared with elemental superconductors, and their superconductivity was thus thought to be unconventional. The anomalies were later suggested to be associated with the structural instability that is common in A15 materials.[2] Neutron scattering experiments on V_3Si and Nb_3Sn revealed considerable softening of high-frequency phonons in these materials. On the basis of these experimental findings, theoretical explanations of superconductivity in A15 systems were developed in terms of the interactions associated with the anomalies in the phonon spectrum.

The assumption of strong electron–phonon coupling in perovskite oxides motivated the discovery of high-T_c superconductivity in lanthanum barium copper oxide (La–Ba–Cu–O). Copper oxides were then named as new high-T_c superconductors. These superconductors again exhibit many interesting properties. The driving force for superconductivity in cuprates seems to go beyond what can be explained by the conventional Bardeen–Cooper–Schrieffer (BCS) theory. Early observations of the negligibly small oxygen isotope exponents in optimally doped $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-\delta}$ argued against phonon-mediated theories and instead in favor of mechanisms that did not involve phonons. However, this was challenged by later experiments on non-optimally doped materials, in which the oxygen isotope exponent was observed to exhibit large variations, sometimes even taking values near unity.

At present, lattice vibrations and excitations of electronic origin such as fluctuations in spin or electric polarizability are considered to be the two most likely candidate mechanisms for Cooper pairing in cuprate superconductors. Carefully designed studies have revealed that both make important contributions to determining the strength of pairing interactions.[3] A similar conclusion of a very sizable electron–phonon interaction was also drawn for FeSe,[4] the most extensively studied iron-based superconductor. A strongly enhanced T_c of a monolayer FeSe film on $SrTiO_3$ was suggested to result from the strong electron–phonon coupling due to a soft mode in the interface. Soft phonon modes and phonon softening have been extensively reported in almost all high-T_c cuprates. For example, a significant softening of the phonon mode was reported in $Bi_2Sr_2CaCu_2O_8$ from ultrasonic measurements, and a soft mode

was observed in $HgBa_2CuO_{4+\delta}$ from inelastic X-ray scattering measurements. Although the degree to which soft modes contribute to superconductivity remains uncertain, their essential role in these cuprates is clear.

The newest high-T_c family are the superhydrides. The highest reported T_c values are those in LaH_{10}: 260 K–280 K[5] and 254 K[6] from resistance measurements and 278 K from magnetic susceptibility.[7] In plots of presently available experimental data, the maximum T_c is found at a pressure of 160 GPa–170 GPa.[7] X-ray diffraction studies have revealed a structural transition from the *Fm*-3*m* structure above 170 GPa to the *R*-3*m* phase below 165 GPa. This result is consistent with the predicted dynamic instability of LaH_{10} below 200 GPa. Systematic studies[8] of superconducting hydrides have revealed that increasing the coupling constant near the instability by phonon softening increases T_c but also drives the instability. The hydrides reach their maximum T_c just before instability. Therefore, the observed maximum T_c at 160 GPa–170 GPa in LaH_{10} is associated with the lattice instability and thus is driven by a soft mode. Similarly, the soft mode may also account for the discontinuity in the variation of T_c with pressure at 225 GPa and its rapid enhancement afterwards up to the record value of 287.7 K at 267 GPa in carbonaceous sulfur hydrides.[9]

III. A POOL OF HIGH-TEMPERATURE SUPERCONDUCTORS

The soft mode scenario described above indicates a possible direction in which to search for high-T_c superconductivity in materials near structural instability. Superhydrides have proved to be among such leading high-T_c materials. In these systems, the structural instability driven by the soft mode is related to the fluctuation and formation of H_2-type units.[8] Enhancement of T_c at lower pressures should be possible if the appropriate element(s) can be found to form hydrides with H_2 units in the lattice.

A connection between ferroelectricity and superconductivity was noticed long ago.[1] Oxide ferroelectrics with large anharmonic properties hold promise as possible high-T_c superconductors. The large anharmonicity often results in low thermal conductivity. If the anharmonicity is dominated by soft mode behavior, then the thermal conductivity will be further reduced upon lattice compression, which favors enhancement of the figure of merit of a thermoelectric material. This happens most often in semimetals. For similar semiconducting thermoelectric materials with narrow bandgaps, a complicated band structure near the Fermi level corresponds to a large Seebeck coefficient. Applying pressure can drive the material to approach the semiconductor–metal boundary or a structural transformation. Highly efficient thermoelectric materials are thus good candidates for exhibiting high-T_c superconductivity under pressure.

Sharing similar material traits with thermoelectrics, topological insulators with their unique electronic structures also provide a pool of materials in which high-T_c superconductivity might be found. Here, pressure is needed as a tool to tune the material to the lattice instability and strong electron–phonon coupling.

Soft modes and the associated order–disorder transitions have also been reported in *p*-oligophenyls. We have found that superconductivity can be induced in such organic molecules by the introduction of charge carriers through chemical doping. T_c values as high as 120 K have been obtained in such systems under ambient pressure and have been confirmed by magnetization, resistance, and angle-resolved photoemission spectroscopy (ARPES) measurements.

IV. EXAMPLE OF AN INTIMATE LINKAGE

Some transition metal dichalcogenides such as 2*H*–$NbSe_2$, 2*H*–TaS_2, and 2*H*–$TaSe_2$ can serve as model systems to observe the soft mode picture and its intimate relation to superconductivity. A broad peak, assigned as the two-phonon mode, is observed in the Raman spectra of these materials. On cooling, the energy of the two-phonon mode decreases (softening), while its spectral width increases and its spectral intensity weakens. This soft mode disappears at the lock-in temperature T_{CDW} of the charge density wave (CDW). Below T_{CDW}, the phonon modes associated with the CDW order appear, and their energies increase with decreasing temperature. These observations reveal the competition between the two-phonon mode and the CDW modes. The latter are a consequence of the former at low temperature. Or, simply speaking, the CDW modes gain energy from the two-phonon mode. On further cooling, the material enters the superconducting state. Below T_c, the CDW modes gradually lose their spectral weight, and the Higgs mode grows as a result. Interestingly, the total spectral weights of the Higgs mode and the CDW modes remain constant at each temperature below T_c. This indicates that the same electron–phonon interaction should account for both the CDW and superconductivity. Energy transfer from the soft mode to the CDW and then to superconductivity can be clearly observed in these transition metal dichalcogenides. With the application of pressure, T_{CDW} decreases while T_c increases. At a critical pressure, the CDW phase melts and T_c reaches a maximum before decreasing slightly owing to the reduction in the interaction strength and the appearance of lattice disorder under heavy compression. The perfect interplay among the soft mode, the CDW, and superconductivity in such low-T_c systems at ambient pressure and at high pressures resembles the phase diagram of many other superconductors, such as the high-T_c cuprates. Thus, a theoretical approach to superconductivity that considers the role of the soft mode in cuprates or other high-T_c materials deserves further development.

V. EXPLORING SUPERCONDUCTIVITY IN HYDROGEN

Early work suggested the existence of a high T_c in metallic hydrogen mainly on the basis of the high value of the Debye temperature. However, the theoretical existence of a soft phonon mode in dense hydrogen was also emphasized.[10] The softness of the transverse phonons can lead to significant enhancement of the coupling constant. Thus, a very high T_c of about 600 K is expected for metallic hydrogen near the lattice instability. Experimental efforts to detect the soft mode as well as the expected very high-temperature superconductivity in metallic hydrogen are therefore highly desirable. Recent developments in high-pressure experimental techniques give grounds for optimism regarding the discovery of metallic hydrogen in the solid state.

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