FIRST CO-ORDINATION SYMMETRY IN THE STRUCTURE OF HIGH CRITICAL TEMPERATURE SUPERCONDUCTORS

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ABSTRACT We propose that the mechanism of superconductivity in high-T_c materials is related to close-packed (icosahedral) symmetry in the co-ordination sphere of the copper atoms, or the atoms of other variable valence elements. The origin of the superconductivity in perovskite-type materials is attributed in part to a three-dimensional nesting of the Fermi-surface with the boundary of a Jones-zone polyhedron, causing a 'partially-gapped' Fermi surface. The pairing mechanism is the result of coherent collective fluctuations that arise from a three-dimensional 'breathing' of distorted perovskite structures, which we associate with icosahedral close-packed-seeking symmetry and with the mixed-valence behaviour of copper atoms.

INTRODUCTION

Many questions concerning the mechanism of superconductivity in high T_c materials are as yet unanswered [1]. Several proposals [2-4] suggest that a non-phonon explanation is needed. It is widely accepted that these materials are metastable, a behaviour associated also with earlier enhanced-T_c materials [5]. There are clear indications, as well, of metastability [6,7] in many recent superconducting ceramics. Mostly, low-dimensional mechanisms of superconductivity are popular [8,9]; however, several recently reported measurements [10,11] and their various interpretations [12-14] have pointed to the reality of a three-dimensional electronic mechanism. We propose a three-dimensional effect arising from close packed Frank-Kasper co-ordination [15] in the recent high-T_c and the earlier reported enhanced-T_c (A15) materials. Our proposal extends - to three dimensions - the interaction between Fermi surface and Jones zone invoked in two dimensions by Mattheis, and later Jorgenson and others [8-9,16-17], with 'nesting' of Jones zone (JZ) and Fermi surface.

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FERMI-SURFACE INTERACTION WITH THE JONES-ZONE POLYHEDRON

Generally, a JZ effect on T_c is well substantiated for metallic superconductors. An enhancement of T_c arises when the Fermi-surface interacts with the JZ [18]. The Jones-zone, whose geometry in k-space is strongly related to the symmetry in real space of the crystalline structure, is a complex polyhedron bounded by a number of planes that is determined by the number of atoms in the elementary cell. The JZ boundaries can be determined experimentally from the first non-vanishing structure factor in x-ray diffraction studies. For complicated crystalline structures the JZ polyhedron is bounded by more and more planes and becomes nearer and nearer to spherical, as is observed for amorphous metallic alloys [19].

Contact of the Fermi surface with the Jones zone over a large part of the effectively spherical JZ surface (ie a 'nesting' of the two) can occur, and an accumulated van Hove singularity results. As a result of this nesting the energy gap produced at the Fermi surface, on interaction with the JZ polyhedron, extends over a large part of the Fermi surface. To surmount the accumulated energy gap a large activation energy is needed, leading to a locally stabilised system. The FS-JZ interaction causes a density-of-states (DOS) enhancement near to E_F, with formation of a pseudogap at E_F. The existence of a pseudogap has been postulated by others [20-23] on different premises and is also a well known characteristic of some metastable phases [24]. The FS-JZ interaction would thus be tuned with the carrier density; which has indeed been observed [25].

On slight shift of the Fermi surface in k-space, within limits set by the JZ inscribed and circumscribed spheres [26], the position and direction in k-space in which the 'partial' gapping occurs will alter but the average partial-gap remains constant and is associated always with the Fermi surface. This produces the partially gapped Fermi surface that Little and others [27-29] have suggested could contribute to high-T_c superconductivity. Indeed, there is much experimental evidence to support the occurance of exciton-related electron pairing [4,30], consistent also with the general observation of metastability in these systems [31].

We propose that these electronic effects, originate from the seeking of close packed Frank-Kasper co-ordination of twelve in these materials. A compact cluster is formed around the copper atoms, seeking a dense atomic arrangement as well as an optimal (spherical) JZ. The most compact cluster is, in fact, icosahedral [15]. In general, we have observed a relation between metastability and icosahedral-seeking symmetry [26], while a relation between symmetry-based metastability and high-T_c has also been proposed [32,33].

We illustrate a possible geometrical rearrangement, of a lattice with co-ordination twelve to icosahedral co-ordination in Figure 1. The concept of icosahedral-seeking symmetry is supported also by the

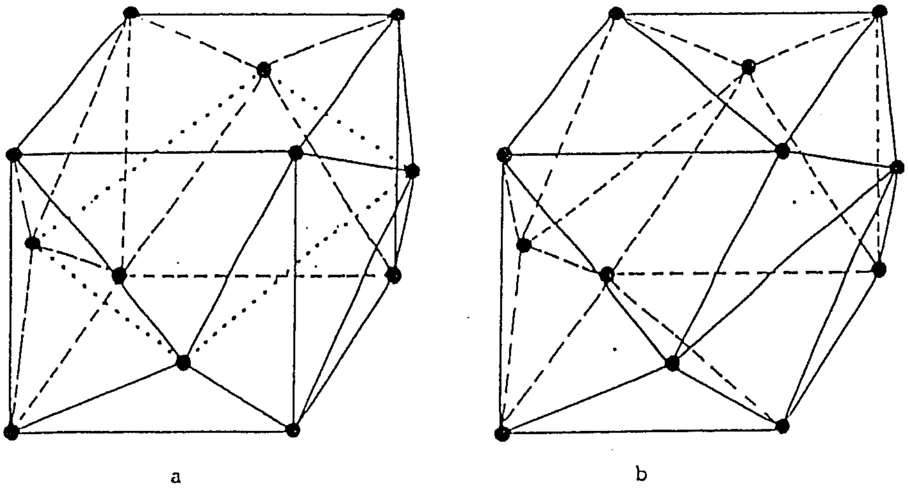

Fig. 1 The icosahedral-seeking nature of a close-packed system with first neighbour co-ordination of 12.

postulate of a quasi-crystalline ground state [34]. The icosahedral-like cluster is associated with a spherical-seeking JZ, which originates from the tendency for structures to form with smallest possible electron-to-atom ratio [e/a].

Turning to the new 'perovskite-type' high-T_c superconductors, we observe (Figure 2a) that their elongated octahedral basic cell has a surrounding structure that has close packed co-ordination (Figure 2b). We note that in all the lanthanum, barium (ReBaCuO), bismuth and thallium (Aurvillius-compound) based high-T_c superconductor structures, the Frank-Kasper co-ordination number is twelve (characteristic of close packing). Since the structures are far from either fcc or hcp (close packed geometries), the displacements needed must be those for which close packing gives a Voronoi first co-ordination polyhedron close to a distorted icosahedral structure (Figure 2c). The proposed displacements would be created in only a very short time, as collective and coherent movements associated with the thermal fluctuations in these materials.

The distorted structure cannot be 'frozen in', because of both its higher ground-state energy and the continuous thermal fluctuations; it is only locally stable, as a result of the lower energy of the special local cluster. The collective fluctuations do not, of course, invoke icosahedral symmetry; any symmetry which causes nesting of the FS and JZ, such as other Voronoi polyhedral co-ordinations, will be more stable.

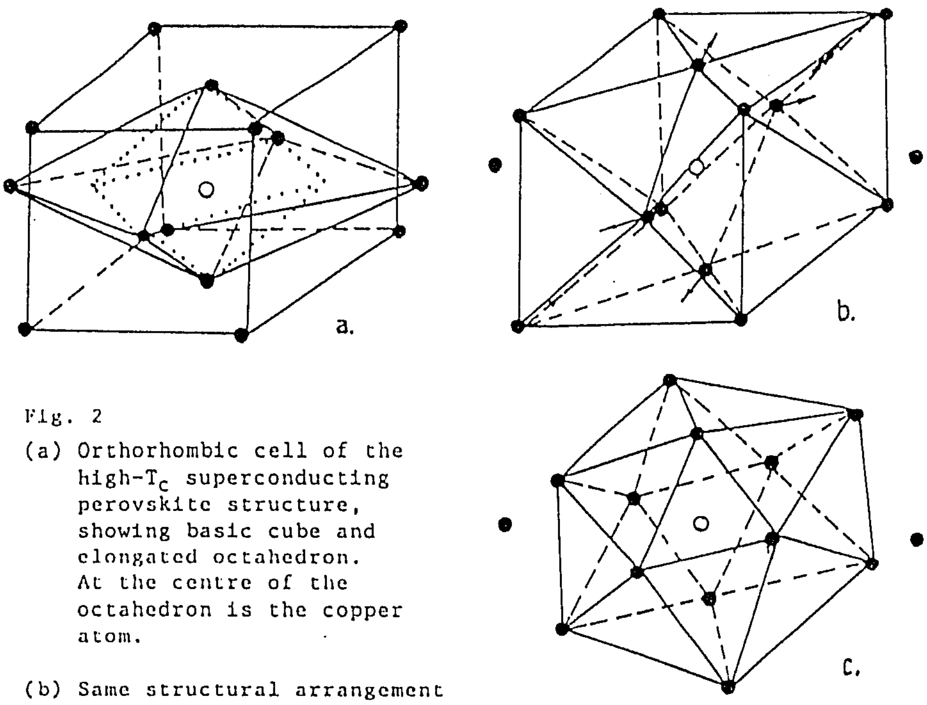

Fig. 2

(a) Orthorhombic cell of the high-T_c superconducting perovskite structure, showing basic cube and elongated octahedron. At the centre of the octahedron is the copper atom.

(b) Same structural arrangement as (a) but highlighting the icosahedral five-fold co-ordination (or bonding) for atoms in the basic cell.

(c) The icosahedral co-ordination 'cell' formed by the displacements indicated in (b).

FS-JZ INTERACTION FOR HIGH-T_c CERAMIC SUPERCONDUCTORS

For insulator-states of recent high-T_c ceramic materials, the FS-JZ interaction can be 'tuned' by hole (T-phase) or electron (T'-phase) doping. We then create a situation in which the first co-ordination sphere prefers a non-frozen-in collective fluctuation, seeking to reach the most stable local clustering arrangement. Random thermal fluctuations, in the effective co-ordination, become collective coherent fluctuations that strive towards the most stable arrangement within the local cluster. The result is a large collective motion in the co-ordination sphere, driven by the electronic structure. This, we believe, is the mechanism responsible for the pairing; promoted by collective, coherent fluctuations. It has been shown [35] that a continuous transformation occurs between crystalline and quasi-crystalline states containing icosahedral clusters. These short-lived clusters, with closest possible packed, Voronoi-type polyhedral first co-ordination, can give rise to high T_c superconductivity by an excitonic mechanism, resulting from the JZ-FS interaction.

Note that the 'distortions', or more precisely the collective fluctuations, exist in single-crystalline materials. No stress or disorder is needed; the behaviour is intrinsic. Indeed, the thermal parameters obtained from neutron diffraction data [36] show extreme fluctuations within the first co-ordination region of the copper atom, which are enhanced below T_c.

Another fact to note is that all the high-T_c superconducting compounds are mixed-valence based systems. In La_2CuO_4, the substitution of La^{3+} by Sr^{2+} converts part of the Cu^{2+} to Cu^{3+}; in $YBa_2Cu_3O_{7-x}$, part of the Cu^{2+} is converted into Cu^{3+} or Cu^+ by varying x, the oxygen-deficiency parameter; and for the recent Bi-compound superconductors, the variability of the Bi-valency is important.

Let us consider a Cu-oxide layer spanned by the Ba-sublattice in YBaCuO. The Cu ions have the Frank-Kasper close packed co-ordination of twelve. The oxygen-vacancy rearrangement, in the vicinity of copper atoms, produces the well-known tetragonal-orthorhombic phase transition. An extra phase transition can be observed in the orthorhombic phase [37] near to the superconductor transition. This second oxygen-vacancy rearrangement produces a structural instability [38], which can easily be understood in terms of the electronic structure [39,40] of these compounds. Some of the Cu^{2+} ions may result from extra holes, so that the oxygen co-ordination of the Cu^{3+} ions will be deformed with respect to that of the Cu^{2+}. The ionic radius of Cu^{3+} becomes smaller, implying that the oxygen octahedron will contract isotropically. If the Cu^{2+} wavefunctions overlap sufficiently with the oxygen wavefunctions, the extra Cu^{3+} ion has high mobility through the lattice. The probability of electron tunnelling is also enhanced by vibronic effects; eg by a 'breathing' deformation of the oxygen octahedron, or by soft optical-phonon assisted charge-carrier hopping, which is a common feature of these compounds. Local deformation causes a long-range layer distortion [32] in the vicinity of the Cu^{2+} ions (Figure 3); this local deformation is consistent with the collective coherent fluctuations referred to above.

The mixed valence mechanism is not unlike the bi-polaronic excitation mechanism proposed [41] for the high-T_c behaviour of these ceramic materials. The large deformability in the neighbourhood of the copper atoms helps the close-packed to non-close-packed 'breathing', with creation of regions of distorted icosahedral co-ordination symmetry and clustering. This breathing mechanism will be readily adopted by the boron compounds which have real and stable icosahedral arrangements [42].

We propose that a three-dimensional charge density wave (CDW) can be produced. This is not the two-dimensional CDW proposed by Mattheiss and discussed by others [43] for the lanthanum based LaMCuO-type superconductors. The Mattheiss calculation, for pure La_2CuO_4, involves a two-dimensional Peierls distortion (or a 'frozen-in' CDW) with either a planar or a two-dimensional quadrupolar 'breathing' of the oxygen atoms. The latter would allow a semiconductor gap to open at E_F and, as described by Stavola et al [43], would destroy the potential superconductivity. The presence of the divalent metal atom (Ba or Sr) prevents the CDW from being frozen in; it essentially stabilizes the metallic phases (by lowering E_F) and, in the Mattheiss model, allows oxygen-atom 'breathing' in two dimensions (ie a warping of the CuO crystal planes) without spoiling the superconductivity. Evidence for a Peierls distortion was found in vibrational spectra by Stavola et al, who proposed that doping with the divalent metal 'tunes' the planar-oxygen breathing distortion. Weber, however, discounts the two-dimensional warping of the CuO plane as being the sole explanation of the Peierls distortion, which again supports our suggestion that a three-dimensional icosahedral warping or breathing is occurring. Other investigations [44,45] also support these ideas.

It is also of note that a Peierls distortion, hitherto accepted only in one-dimensional and two-dimensional models, has recently been observed experimentally in the p-bonding of liquid arsenic [46]. Here it must be a three-dimensional effect, since this is an amorphous state, supporting our contention that three-dimensional breathing distortions can occur in the solid, located in the layer containing the Cu-O planes. The displacements would bring about a close packed tetrahedral arrangement, producing microscopic short-lived coherent fluctuations in Voronoi co-ordination regions, with tetrahedral Frank-Kasper packing, which have been observed to have an important role in the stabilization of the metastable systems [26,35]. A 'non-frozen-in' or gliding CDW in three dimensions can result from a 'breathing' of microscopic close-packed cluster regions. In the crystal, periodically occurring distorted (icosahedral-like) dense Voronoi-polyhedral co-ordination regions would allow for the complete filling of real space needed.

The 'breathing' mechanisms is the same as described for valence-changes (see Figure 3). We illustrate schematically (Figure 4) a mode in which this short-range collective breathing can occur throughout the whole system. The oxygen atoms, as in the Mattheis model, are primarily involved; their displacement (for rotation of CuO octahedra, as needed for distorted icosahedral-cluster packing) has in fact been observed by Onoda et al [47] in crystal structure determinations of LaMCuO-type superconductors. The solid-line regions (of Figure 1c) are the momentarily more stable arrangement, ie neighbourhood regions linked in 3-dimensions (for example through corner atoms, as here) to the next-neighbour stabilized micro-region. This example illustrates a 'commensurate' breathing in the layers containing the Cu-O planes; complex 'non-commensurate' forms of breathing are also possible, in which the close packed regions (highlighted in Figure 4) actually overlap. The incommensurate structure has in fact been found in a high-T_c system [48], and a transition induced by a co-operative Jahn-Teller effect is also possible between the commensurate and incom-

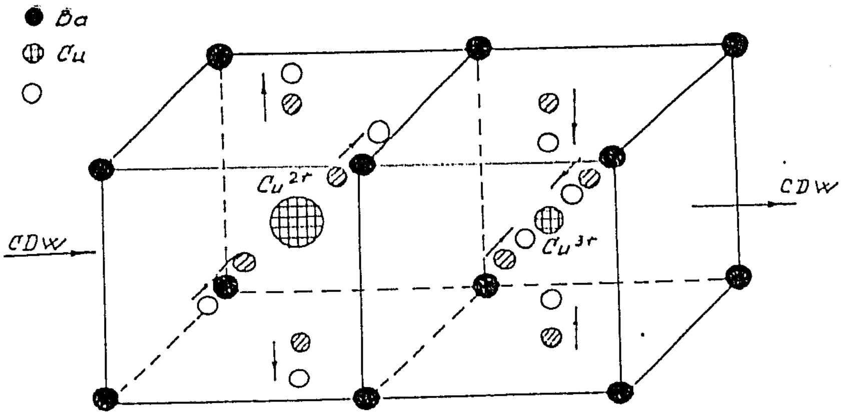

Fig. 3 Changes in the oxygen-environment around Cu-ions of different valency.

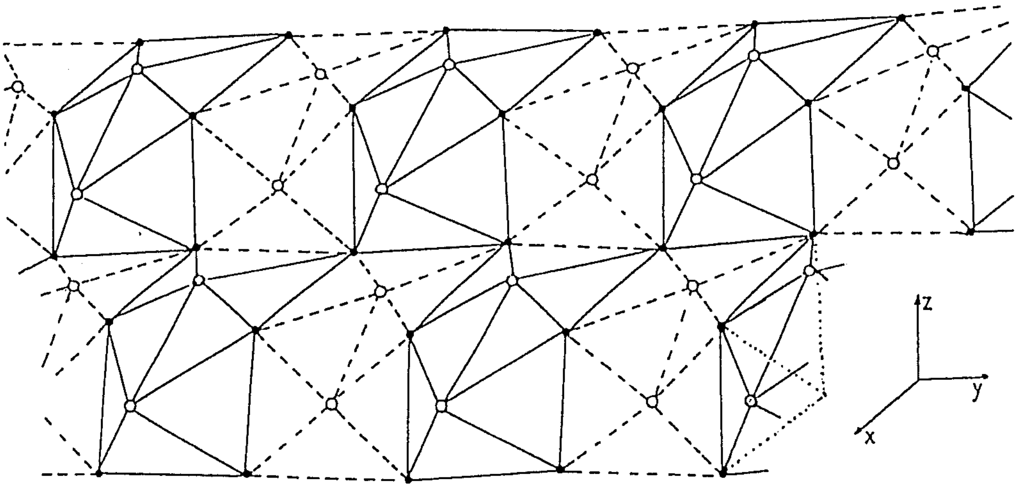

Fig. 4 A schematic of the proposed 3-dimensional breathing displacements of oxygen atoms in perovskite-type materials, from icosahedral local micro-regions (solid-lines) to non-icosahedral micro-regions (dotted lines) : o - oxygen atoms : • - Ba,La,Y atoms. Note that a two-dimensional planar warping, proposed previously by Mattheiss and others [3,8-13] is apparent.

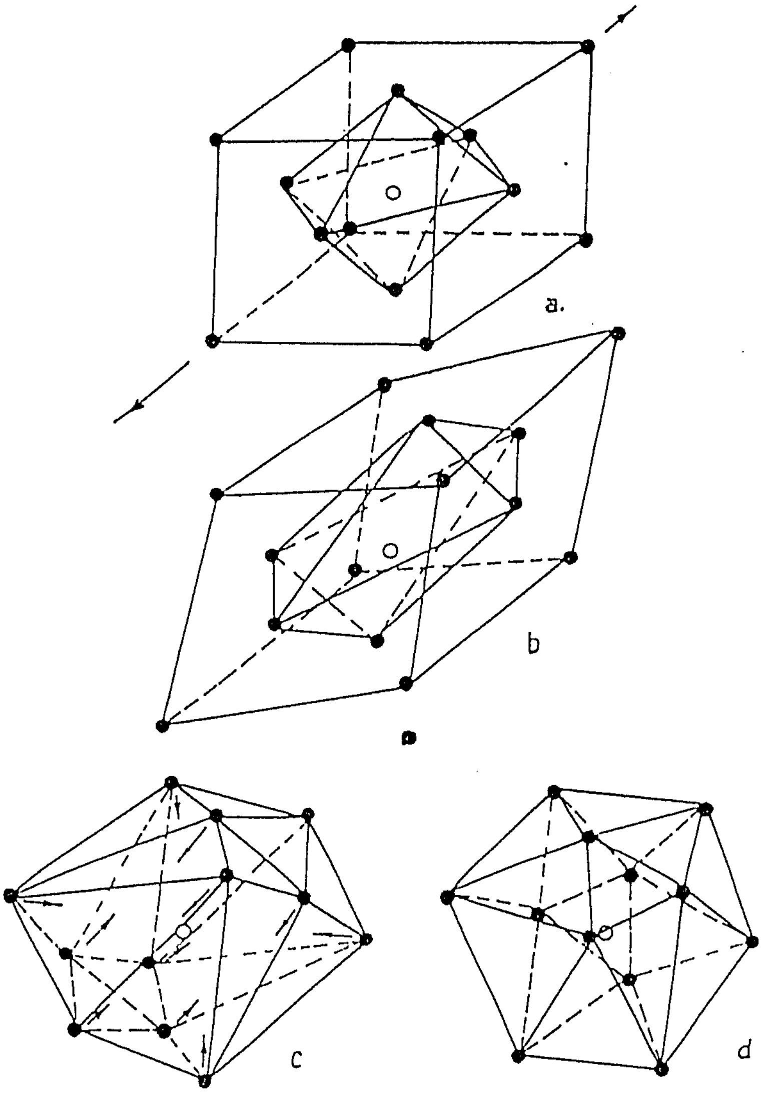

Fig. 5 Distortions of the Frank-Kasper co-ordination polyhedra in a mechanically stressed distorted perovskite structure.

(a) Non-distorted perovskite.

(b) Perovskite structure diagonally elongated by external mechanical stresses.

(c) The distorted Frank-Kasper co-ordination polyhedra, now a distorted icosahedron.

(d) Distorted icosahedron.

mensurate structures [49,50]. This is in good agreement with the models based on the highly anharmonic oxygen vibrations insuperconducting oxides [51], and is well supported by the role of the shear distortions [52]. We note, that the CDW's are located in the thick layers containing the Cu-O planes and are not extended into the whole elementary cell. The three-dimensionality covers changes in the three-dimensional neighbourhood for these layers. Figure 4,. for example, shows the case of an Aurvillius-phase containing double CuO planes in one cell. An experiment to test for the existence of sliding CDW's has been proposed [53], involving nuclear magnetic resonance measurements. The theoretical basis of CDW de-pinning, by electron tunnelling, has also been described [54].

According to these concepts, the pair-creation is not phonon-induced, but is caused by other collective lattice vibrations, involving large clusters of first co-ordination (variable-valence) atoms. This causes the isotope effect not to follow the phononic $T \propto M^{-\frac{1}{2}}$ relation, because of the large mass of the collectively moving clusters. It becomes $T \propto M^{-\alpha}$ where $\alpha < \frac{1}{2}$. Indeed, we observe $\alpha = \frac{1}{2}$ (BCS model) only for simple metals and for Ba(PbBi)O , while for the A15 alloys and other transition metals and alloys, including their amorphous forms, as well as for the recent high-T_c ceramics, $\alpha < \frac{1}{2}$.

The icosahedral-like close packing also causes special effects near to the Fermi-energy (E_F) [26,35]. As described above, the spherical-seeking JZ causes a pseudogap to appear at E_F, similar to the electronic structure effect observed for amorphous metallic systems. The electronic theory of superconducting glasses becomes applicable [55,56]. A relative enhancement of N(E) in the vicinity of E_F has in fact been observed experimentally for high-T_c materials, by both UPS [57] and XPS [58-60], and has been interpreted from measurements of their specific heat [61] as well as their thermo-electric power [62]. The pseudogap has also been deduced [20-22] from theoretical considerations, in an analog of the Nagel-Tauc theory [24] for metallic glasses; while the glass-behaviour has been measured [63-64], and computer simulated [65].

The special properties of the optimum $YBa_2Cu_3 O_{7-x}$ composition, compared to other stoichiometries, have been shown by a combination of differential thermo-analysis (DTA) and soft x-ray emission spectroscopy (SXES) [66]. The highest T_c is exhibited by the most metastable state of the material. The SXES data is consistent [67] with clustering in the first co-ordination sphere of copper. Other supporting evidence can be found in the enhanced T_c for finite-size fractal aggregates [68], and in current network modelling of superconductive transport [69,70], in which icosahedral-seeking geometry is easily recognised. The three-dimensional behaviour is observed in specific-heat measurements [10], while it can be seen easily for the copper-free new perovskite superconductors [71,72].

Supporting evidence for the so-called 'partial gapping' can be found in far infra-red measurements of the superconductor energy gap, showing the latter to be highly anisotropic [73] and to vary with direction in k-space; while its temperature dependence - characterizing the Peierls instability - remains BCS in 'functional form' [74,75]. Additional support includes the isotope effect being anomalously small for oxygen and non-existing for copper [76].

Lastly, we note that the new high-T_c materials generally have highly distorted extended grain-boundary structures which in many cases are amorphous. The disorder in these is localised to the grain boundary, where our proposed short-range icosahedral co-ordination can even more readily occur. The observed crystal-twinning in the new high-T_c perovskite-type superconductors [77] is indicative of intra-granular distortion of the basic 'perovskite' cube, giving a structure that will inevitably be closer to icosahedral. We show in Figure 5 an example of one highly distorted perovskite structure, with its icosahedral-like Frank-Kasper co-ordination clearly distinguishable.

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