High-temperature superconductivity (a review of theoretical ideas)*

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The topic of high-temperature superconductivity began first to be seriously discussed starting in 1964, after it was hypothesized that in quasi-one-dimensional polymer chains[1] and quasi-two-dimensional systems (the interface of a metal-insulator sandwich, etc.)[2] the critical temperature T_c could be increased substantially as a result of an exciton mechanism of attraction between electrons. The state of this subject up to 1977 has been detailed in a collective monograph[3] (see also the more recent reviews in Refs. 4 and 5).

The discovery of metal-oxide ceramics with a high critical temperature[6] proved to be a surprise and was not foreseen by the theoreticians. This discovery has confronted theory with all the more serious problems. It still remains to establish what the mechanism of the superconducting pairing is in the metal-oxide ceramic material (determining first of all whether it is the ordinary phonon mechanism, or if it is one of a number of "exotic" mechanisms), to find if there are any materials with a high T_c other than the metal-oxide ceramic compounds, and to assess the prospects for a further increase in this quantity.

Unfortunately, experimental investigations of metal-oxide ceramics have in fact only just begun, and the results are still far from complete and even partially contradictory. Hence, the initial data required to solve the problems enumerated above are still lacking. This is also true as regards the possibility of evaluating the models that have already been proposed for the superconductivity of metal-oxide ceramics. In such a situation this paper cannot claim to be anything more than a short review of the existing ideas and suggestions regarding the possibility of raising the critical temperature of superconductors.

1. The key to the problem of high-temperature superconductivity is the question of the dependence of the critical temperature on the characteristics of the material. In the Bardeen-Cooper-Schrieffer (BCS) theory the quantity T_c is connected with the width $k_B\theta$ of the energy region of attraction around the Fermi level, with the density of electron states $N(0)$ at the Fermi level, and with the matrix element V of the interaction potential

$$T_c = \theta \exp(-\lambda_{\rm eff}^{-1}), \quad \lambda_{\rm eff} = N(0)V. \tag{1}$$

For the phonon mechanism of superconductivity, which corresponds to the attraction between electrons via their interaction with lattice vibrations, the quantity θ in (1) has the sense of a Debye temperature θ_D.

More realistic theories (the theory of Èliashberg, the weak coupling theory for media with an arbitrary dispersion of the dielectric function) reduce to a refinement or a modification of formula (1), the derivation of which was based on a number of simplifying assumptions. The results depend strongly on the value of λ—the dimensionless coupling constant for electrons with lattice vibrations.

In the case of weak coupling ($\lambda \ll 1$) it is necessary to substitute into formula (1)

$$\lambda_{\rm eff} = \lambda - \mu^* = \lambda - \mu\left(1 + \mu \ln \frac{E_F}{k_B\theta}\right)^{-1} \tag{2}$$

($\mu \lesssim 0.5$ is the dimensionless coupling constant of the Coulomb interaction, and E_F is the Fermi energy), taking θ to mean the logarithmic average of the phonon frequency (see Ref. 3, Ch. 2). As θ increases T_c goes through a maximum that becomes lower as λ decreases. One finds in the literature the inequality $\lambda \leqslant \mu$, which, if it were necessarily valid, would place an absolute lower limit on T_c. This inequality comes from the expression for the interaction through the longitudinal dielectric function $\varepsilon(\omega,k)$ of the medium

$$\mu - \lambda = 4\pi e^2 N(0) \left\langle \frac{1}{k^2\varepsilon(0,\ k)} \right\rangle$$

(the brackets denote an average over the momenta $\hbar k$) and from the condition of stability of the medium, which takes the form $\varepsilon(0,k) \geqslant 1$ (for $k \neq 0$). Actually, however, the stability condition has the form $1/\varepsilon(0,k) \leqslant 1$ (for $k \neq 0$). This permits values of $\varepsilon(0,k) < 0$ and of $\lambda > \mu$ and thereby lifts the limitation on T_c mentioned above (see Refs. 3 and 7). Such values not only are possible in principle, but are in fact realized in many media[4,5] [for instance in the alloy PbBi, $\lambda \approx 2.6 > \mu$ (see Refs. 5 and 8)].

In the case of intermediate coupling ($\lambda \sim 1$) there are a number of formulas for T_c similar to (1) having a semiempirical or model-dependent nature, and in any case having an unknown or a clearly narrow region of application.[3,5,8] The simplest of these corresponds to replacing (2) by

$$\lambda_{\rm eff} = (\lambda - \mu^*)(1 + \lambda)^{-1}. \tag{3}$$

In the case of strong coupling ($\lambda \gg 1$) the equations of Èliashberg give

$$T_c \approx \frac{\hbar\bar{\omega}}{k_B} \lambda^{1/2}, \tag{4}$$

where $\bar{\omega}$ is some average phonon frequency (see Refs. 9 and 10, and Ref. 3, Ch. 4). It is important to note that for $\lambda \gtrsim 1$ the quantities θ and λ can no longer be considered independent: an increase in λ brings about a softening of the phonons, and a decrease of θ due to the screening action of the conduction electrons. This reverse effect of the electrons on the lattice is described (in the simplest case of a single peak in the density of phonon states) by the expression

$$k_B\theta_D \approx \hbar\omega_0 (1 + c\lambda)^{-1}, \tag{5}$$

where ω_0 is the bare ("pseudoatomic") frequency and c is a constant of the order of unity. The value of $\hbar\omega_0/k_B > \theta_D$ places an upper limit on the critical temperature in the case of weak coupling (see Refs. 5 and 10, and the references cited therein).

The content of this section applies not only to the phonon mechanism, but in a larger measure also to the exciton mechanism of superconductivity (see below).

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2. From the discussion above it follows that there are in principle no prohibitions on a significant increase of T_c within the framework of the phonon mechanism for superconductors which have sufficiently large values of θ and λ. Rough estimates using the formulas above demonstrate this fact. For instance, according to (1), the values $\lambda_{\text{eff}} \lesssim 3$ and $\theta \sim 300$ K give $T_c \sim 100$ K and formula (3), with $\mu^* = 0.1$, $\theta \sim 300$ K and $\lambda = 1$ and 3 gives $T_c \sim 30$ K and 75 K, respectively. These values are similar to those given by the other formulas mentioned above for the intermediate-coupling case. The whole question reduces to whether or not there exist superconducting materials for which the values of both θ and λ are sufficiently large at the same time.

Until 1986 such materials were unknown. Many metals have high values of $\theta = \theta_D$ of the order of 400 to 500 K, and even 1400 K for beryllium[11] (high values of θ_D were also expected for organic superconductors containing the light atoms H or C).[12] A number of superconductors have large values of $\lambda \sim 1$–2. In these materials, however, a large θ_D has inevitably been accompanied by a small λ and vice versa [this situation entailed for the known superconductors the inequality $T_c \lesssim 25$ K, which corresponds in formula (1) to, e.g., $\theta_D \lesssim 500$ K and $\lambda_{\text{eff}} \lesssim 1/3$]. It is for this reason that published estimates for the phonon mechanism have been very conservative ($T_c \lesssim 30$–40 K; see, e.g., Ref. 3, Ch. 1). It was, however, always stipulated that this conclusion could not be considered absolute, since it was based on rough estimates and the extremely limited experimental data known at that time (in particular, the case of metallic hydrogen, with $\theta_D \sim 3000$ K and $T_c \sim 100$–300 K has been discussed; see Refs. 3 and 5, and references cited therein). The possibility in principle of a relatively high T_c within the framework of the phonon mechanism for not too exotic materials has always been conceded, but it has not been considered very probable.

3. It is entirely possible that with the discovery of metal-oxide ceramic superconductors with their high critical temperature (for a review of experiment see Ref. 6) the situation in the aspect considered has changed. Although, as has been emphasized, data required for a complete solution of the issue of the superconducting mechanism of the metal-oxide ceramics are not yet available, nonetheless it is possible at this time to discuss the possibility that it is just in the metal-oxide ceramics that the fortunate combination of relatively high Debye temperatures θ_D and large electron-phonon coupling constants λ has been produced for the first time, and that the ordinary phonon mechanism of superconductivity is responsible for the high critical temperature for these compounds.

There are a number of serious arguments in favor of this possibility, having to do mainly with the metal-oxide ceramic La–Sr–Cu–O and those similar to it, with $T_c \simeq 36$ K (see Refs. 13–15 and also Refs. 16 and 17, in which the electronic structure of the tetragonal crystal La_2CuO_4 is calculated). The distinctive character of these compounds in the context of the present interest is connected with the fact that they have characteristic "stiff" optical lattice vibrational modes [corresponding to large values of ω_0; see formula (5)] which are rather strongly coupled to the conduction electrons. This "stiffness" is due to the small mass of the O ion and the substantial ionic component in the bond, to which about 10 electrons in the unit cell contribute, while at the same time there are only 1–2 conduction electrons, a situation that leads to a small fractional metallic bond and produces a relatively small reverse effect on the lattice (see Sec. 1, above, and also Ref. 3, p. 195). All these properties also lead to a value of θ_D of the order of hundreds of kelvins in combination with a value $\lambda \gtrsim 2$, which is wholly compatible with $T_c \sim 40$ K. This argument is supported by the direct calculations mentioned above, and also by a comparison with the previously obtained metal-oxide ceramic Ba–Pb–Bi–O, with $T_c \approx 13$ K (Ref. 18).

Of course it is still not possible to claim that the nature of the superconductivity in La–Sr(Ba)–Cu–O has been determined. This observation applies even more to the Y–Ba–Cu–O metal-oxide ceramic with $T_c \sim 100$ K. However, the possibility that the high-temperature superconductivity of these compounds can be explained by the phonon mechanism can in no way be excluded. A confirmation of this possibility by subsequent experiments would mean that the solution of the high-temperature superconductivity problem has waited for us, so to speak, on a trivial path.

4. It is not known whether this possibility (the phonon mechanism) is operative in the metal-oxide ceramics with $T_c \sim 30$–100 K. Moreover, one cannot dismiss the possibility that the metal-oxide ceramic is only the first member of a family of high-temperature superconductors that also includes materials with other pairing mechanisms. Therefore, without any claim to completeness, a short review of non-standard superconductivity mechanisms is presented below. In the existing state of experimental investigations of the metal-oxide ceramics, this review must be regarded only as an illustration of the possibilities which will have to be seriously discussed in case the phonon mechanism for high T_c is found to be clearly unacceptable.

The very simple BCS formula (1) written in the form $T_c = \theta \exp(-1/N(0)V)$ already allows us to find the principal factors that bring about an increase in T_c. These are, obviously, increases in, respectively, the width of the characteristic interaction region $k_B T$, the density of electron states at the Fermi level, $N(0)$, and the interaction V. Therefore the discussion below will concern examples of three classes of mechanisms corresponding to these three factors.

Let us begin with a mechanism that leads to an increase in the density of levels $N(0)$. In the standard theory of superconductivity this quantity is the smooth, slowly varying density of levels $N(E)$ at $E = E_F$. The situation will be different if at a temperature close to T_c there is a structural phase transition in which an insulating type of energy gap appears in the electron energy spectrum. Such a gap can be produced by a congruence (nesting) of individual parts of the Fermi surface; that is, a coincidence of these parts of the Fermi surface upon a shift of the electron momentum by a certain amount and a reflection. Near the edge of the gap the density of levels increases and varies rapidly as $N(E) \sim \varepsilon^{-1/2}$, where ε is the distance from the edge of the gap in the allowed region (physically this is associated with the "expulsion" of levels from the gap itself). Now if the electron Fermi level of a doped semiconductor lies in the gap and just at the edge of it, then subsequent superconducting pairing will occur under conditions of a large density of states (and one that varies rapidly with energy), and this also leads to an increase in the critical temperature T_c relative to the case where there is no insulating gap (see Refs. 19 and 20, and Ref. 3, Ch. 5).

The other factor—an increase in the interelectron attraction V—is operative in the bipolaron model of superconductivity. A sufficiently strong electron-phonon interaction will convert the fairly wide conduction band to a narrow polaron band, to which corresponds a large polaron-carrier mass. This same interaction creates bound pairs—bipolarons—with a large binding energy and a small radius (roughly speaking, this means that the two electrons are in a common polarization well). The collection of bipolarons, which are Bose particles, may undergo Bose-Einstein condensation, the temperature of which for a low bipolaron concentration n is given by the usual formula $T_c = 3.31\,\hbar^2 n^{2/3}/k_B m^*$, where m^* is the bipolaron mass. This temperature is also the superconducting transition temperature in this model. For $n \sim 10^{21}$–10^{22} cm^{-3}, and $m^* \sim 10^2 m$ (where m is the mass of the free electron), $T_c \sim 30$–100 K. It should be emphasized that in contrast to Cooper pairs, which overlap in space and condense cooperatively, the bipolarons of small radius condense singly, and in the model considered here there is no energy gap in the usual sense (with the BCS theory in mind).

It is interesting to note that such a superconducting mechanism began to be discussed even before the BCS theory was developed. For instance it was noted[21] that a Bose-Einstein gas of charged particles (particles with spin 0, let us say) would behave as a superconductor. On the basis of the same observation it was proposed[22] that the electrons of a metal "stick together" in pairs (quasimolecules) with a charge $2e$ and spin 0, and the condensation of these pairs also leads to superconductivity. The bipolarons assume the role of the quasimolecules in the bipolaron model (see Ref. 23, where there is a comprehensive bibliography).

Similar in a certain sense to the bipolaron model is the "resonating valence bond" model, which has recently been proposed to explain the superconductivity of the metal-oxide ceramics.[24] In this model the formation of bound pairs of electrons is due not to the electron-phonon interaction but to exchange forces of the Heitler-London type, where the critical temperature scale is determined by the characteristic temperature of the antiferromagnetic transition and can be of the order of 100 K.

5. According to the outline of this paper it remains to consider the third way of raising the critical temperature T_c—widening the region of the attraction; that is, increasing the temperature θ in (1). This topic includes the "exciton" superconductivity mechanism, which has been close to the source of the high-temperature superconductivity problem.[1,2] The idea of this mechanism is that an attraction between conduction electrons can be generated by their interaction not only with lattice vibrations (the phonon mechanism), but also with electrons themselves. In other words, the source of this attraction may be an exchange not only of lattice excitations (phonons) but also of excitations of an electronic type (excitons, let us say). In this case we are not concerned with excitations in a system of conduction electrons [the corresponding excitations are plasmons, and although they give rise to an attraction, it is exhausted by screening of the Coulomb interaction and affects only the value of μ; see formula (2)]; instead the issue is excitations of other electrons, distinct from the conduction electrons. These "other" electrons may belong to deeper bands ("bound" electrons), to nonconducting portions of a nonuniform superconductor, etc.

An important point is that the characteristic energy scale of the electronic excitations, and therefore the corresponding width of the region of attraction near the Fermi boundary $k_B\theta_e$, is much greater than the Debye energy $k_B\theta_D$. This is because, in contrast to the phonon mechanism, here the oscillations are not of a heavy ion, but of a light electron. The relation between θ_e and θ_D can be easily seen from considerations associated with the isotope effect, $\theta_D \propto M^{-1/2}$, where M is the mass of the ion. Replacing in this relation M by the electron mass m, we find that $\theta_e \sim (M/m)^{1/2}\theta_D \lesssim 100\,\theta_D$. From experiments it is also known, of course, that there exist electronic excitons with $\theta_e \sim 10^3$–10^4 K. Substitution of θ_e for θ_D in the BCS formula (1) and in other formulas for the phonon mechanism (see Sec. 1), constitutes the idea of the exciton mechanism.

Of course, for the exciton mechanism to be effective it is necessary that an increase in the preexponential factor θ in (1) and in other similar expressions is not accompanied by a decrease in $\lambda_{\rm eff}$. General considerations admit of such a possibility, but under the necessary condition $\varepsilon(0,k) < 0$ (see Sec. 1 and Ref. 3). In the case of the exciton mechanism the issue must be the negative sign of the electronic part of the dielectric function $\varepsilon_{\rm el}(0,k) = \varepsilon(0,k) - \varepsilon_{\rm ph}(0,k) + 1$, where $\varepsilon_{\rm ph}$ is the contribution of the lattice. It is not easy to satisfy this condition in a real uniform metal, since it puts the stability of the lattice in jeopardy: this can be seen within the framework of the very simple "jellium" model, in which the square of the phonon frequency is equal to the ratio of the square of the plasma frequency to $\varepsilon_{\rm el}(0,k)$, and consequently for $\varepsilon_{\rm el}(0,k) < 0$, the square of the phonon frequency becomes negative. In any case, a consistent theory of the exciton mechanism, which is still lacking, will require that the exchange and correlation interaction (in particular, local field effects) be taken into account; it will require going beyond the weak-coupling approximation, and so forth.[3,5,25]

In general, the prospects of an exciton mechanism from the point of view of "high-grade" theory are unclear. As to various heuristic arguments and estimates, there has not been very much new in recent years (see Refs. 1, 2, 5, 25–28, and Ch. 1, 4, 6–8 in Ref. 3). As before, it appears difficult to expect an effective exciton mechanism for highly conducting typical metals with a uniform structure, and preference should be given to quasi-one-dimensional and especially quasi-two-dimensional structures (destructive fluctuations play a lesser role in the latter), to interphase boundaries, "sandwiches," layered compounds, and so forth.[2,3,26,28] However, the thickness of the conducting layer in these structures must not greatly exceed atomic dimensions.

In the literature there are a large number of superconductivity models which take into account one or another specific interaction of the conduction electrons with the lattice or with "bound" electrons. An analysis of these models (see Ref. 3, p. 201 of the Russian original, and Refs. 18, 27, 29, 30, and 39) lies outside the scope of this paper.

6. It should be emphasized once again that in giving a catalog of nonstandard superconductivity mechanisms the intention is not to make a direct comparison with the properties of the metal-oxide ceramics, although it is now possible to point out a number of features of the metal-oxide ceramics that favor the action of these mechanisms. Among these are, for instance, the layered, quasi-two-dimensional perovskite structure and the large number of collective electronic excitations in the metal-oxide ceramic (the exciton mechanism), the presence of nesting (the dielectrization mechanism and, more indirectly, the bipolaron mechanism), and others.

Furthermore, for the solution of the principal problem—the correct determination of the nature of high-temperature superconductivity in the metal-oxide ceramics—there is still an acute lack of initial experimental data. Suffice it to say that strictly speaking we do not know whether the high-temperature properties are inherent in the ideal crystal of the metal oxide, or whether an important role is played by its imperfections, which are present to excess in the metal-oxide ceramics.

To determine the nature of the high-temperature superconductivity of metal-oxide ceramics (and first of all to resolve the question of whether it reduces to a phonon mechanism) broad investigations of the characteristics of these compounds in the superconducting and normal state, and also the appropriate calculations, are needed. Of special importance from this point of view are studies of single crystals (the role of structural imperfections), x-ray structural measurements (the lattice type), optical measurements (band structure), tunneling spectroscopy and neutron measurements (the phonon spectrum and the electron-phonon interaction), specific heat measurements (the Debye temperature, the density of states, etc.), magnetic measurements (the susceptibility, the critical field, and the penetration depth), tunneling measurements (the energy gap), and others. Calculations should cast light on the band structure and the phonon characteristics (including the electron-phonon interaction) and so forth.

An important measurement for the elucidation of the superconductivity mechanism of the metal-oxide ceramics (although probably very difficult because of their complicated composition) would be measurement of the isotope effect, which, if present, would be indicative of the phonon mechanism. Important information would be obtained by measuring the ratio of the energy gap to T_c, the discontinuity in the specific heat at the transition point, the argument of the exponential in the specific heat at low temperatures, and the like: a comparison of these quantities with the BCS theory could indicate strong coupling, the width of the energy region of the interaction and so forth.

Whatever the true mechanism of the superconductivity of the metal-oxide ceramics may prove to be, it must apparently be characterized by a certain "roughness," a relative insensitivity to the composition, the methods of preparation, and so forth. This is indicated by the relative ease with which the experimental results on the metal-oxide ceramics are reproduced in the different laboratories of the world, and also by the stability of these results.

All the high-temperature superconductors that have been discovered (with the exception of Nb–Ge–Al–O films, with $T_c \approx 44$ K; see Ref. 31) are among the metal-oxide ceramics. Needless to say we cannot dismiss the possibility that there exist other classes of high-temperature superconductors, that can be sought in a systematic way after the nature of the superconductivity of the metal-oxide ceramics has been determined. In this connection we cannot forget the possibilities associated with layered and organic superconductors, "sandwiches," etc.

7. The discovery of high-temperature superconductivity, which already permits operation at liqud nitrogen temperatures, is an outstanding event which may have difficult-to-predict consequences of a scientific, technological and even social nature. It is significant that this discovery did not come about as the result of a systematic and deliberate accumulation of knowledge, but rather it happened more or less accidentally. The time of the discovery, the end of 1986, may also be considered accidental. It could easily have happened at least eight or ten years earlier. This assertion can be attested to by the following astonishing fact: at the end of 1978 a paper was submitted for publication, in which workers at the N. S. Kurnakov Institute of General and Inorganic Chemistry, Academy of Sciences of the USSR, reported the synthesis of various metal-oxide ceramics including the compound La–Sr(Ba)–Cu–O of just the correct quantitative composition; they also reported measurements of the electrical conductivity of these compounds, but not at low temperatures!

We can gain some lessons from the history of the discovery of high-temperature superconductivity.

1) If the mechanism of the superconductivity of the metal-oxide ceramics turns out to be simply a phonon mechanism, this means that the theoreticians have not studied a number of possibilities (in particular, that of a superconductor that has large values of θ_D and λ simultaneously).

2) Experimenters have not been motivated in the direction of high-temperature superconductivity, and there has not been enough communication between low-temperature physicists, chemists, and materials scientists. The very best evidence for this is the fact, discussed above, regarding the publication of Ref. 32. The general atmosphere that prevailed here was a cool attitude towards the problem of high-temperature superconductivity, a topic which occupied only a small number of enthusiasts in spite of its obvious great importance.

3) In the light of the discovery of high-temperature superconductivity we should return to the experiments of 1978 in which a diamagnetic anomaly in CuCl at temperatures of the order of 150–200 K was observed[33] (later a similar effect was found in CdS; see Ref. 34). Unfortunately, as frequently happens, the results were not reproducible (the term "irreproducible superconductors" was even proposed[35]) and it was generally believed that the results were erroneous. Meanwhile the possibility cannot be ignored, especially in the light of the results on the metal-oxide ceramics, that in these experiments high-temperature superconductivity did make an appearance (or else an extremely interesting "superdiamagnetism"[25,36]). If the hypothesis that CuCl and CdS are high-temperature superconductors is really confirmed, but over the period of nine years this has not been understood and the work has been neglected, then another instructive page (and one might say, a sad one) has been written in the history of physics. It is to be hoped that these and other lessons from the history of the discovery of superconductivity in the metal-oxide ceramics will be taken to heart and a program involving a wide and unprejudiced search, such as has been proposed previously,[3,26,37,38] will be undertaken.

Until 1987 the idea of producing materials that remain superconducting in liquid nitrogen remained a dream. Now it is a reality. Now the dream in the field of superconductivity is the creation of room temperature superconductors. We know of no prohibitions on the way to this goal, and values of

$T_c \sim 300$–500 K are in principle attainable. Of course, it is not possible to give any guarantee of this. The experience of the past, however, justifies concluding this paper with an appeal not to repeat old mistakes, not to exhibit excessive skepticism, but to search persistently on a broad front for new high-temperature superconductors.

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