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Phase diagrams of oxygen ordering in high-temperature superconductors $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$

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In the framework of the lattice-gas model the investigation of various possible forms of phase diagrams of $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ (R =rare earth) superconductors is carried out. The calculations are performed by the modified cluster-variation method. The conditions are analyzed which lead to the occurrence of a two-phase region at low temperatures. The problems of $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ system behavior are discussed when the values of the interaction parameters depend on the amount of oxygen. It is shown that at specific conditions imposed on the interaction parameters the region of tetragonal-phase existence can significantly increase at relatively low temperatures. The orthorhombic phase with the reduced wave vector $(\frac{1}{3},0,0)$ is found to occur when the interactions between the next-nearest O—Cu—O chains are considered.

I. INTRODUCTION

The structure and properties of high-T_c superconductors $R\mathrm{Ba}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ (1:2:3) with R =Y , La, Gd, Er, etc., depend on the arrangement of oxygen atoms in basal CuO planes. The oxygens are disordered in the tetragonal (TG) phase and ordered in O—Cu—O chains in the superconducting orthorhombic (O_i) phases. These phases are found to occur at various values of temperature T and oxygen concentration of the CuO plane $c=(1-x)/2$. The ground states of the O_i phases are shown in Fig. 1.

The (T,c) diagram of $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ has been investigated better than in any other 1:2:3 superconductor. The existence of the triple point,[1,2] the location of the TG and O_I boundaries,[3,4] and the changes of orthorhombic structures at $c>0.25$ (Ref. 5-7) are now recognized. However, the interpretation of other data on the structural transitions in $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ still remains ambiguous. This is true in particular when one considers the problems related with the TG and O_{II} phase boundaries at $c<0.25$ (Refs. 2 and 6), the occurrence of two-phase regions at low T (Ref. 8) and of various additional orthorhombic superstructures (see $\mathrm{O}_{\mathrm{III}}$ in Fig. 1) at $c>0.25$.[6,7] Other 1:2:3 compounds are investigated less, but presumably the (T,c) diagram of each 1:2:3 superconductor ought to be qualitatively similar and could be described by a common model. Certainly, the indication of the TG-O_I transition temperature dependence on the ionic radius of the rare-earth atom[9] and the difference of slope of the TG-O_I phase boundary lines in $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ (Ref. 4) and $\mathrm{ErBa}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$ (Ref. 10) suggest some quantitative variation. Therefore the analysis of the possible forms of the (T,c) diagram, performed by variation of interaction potentials, appears to be particularly important.

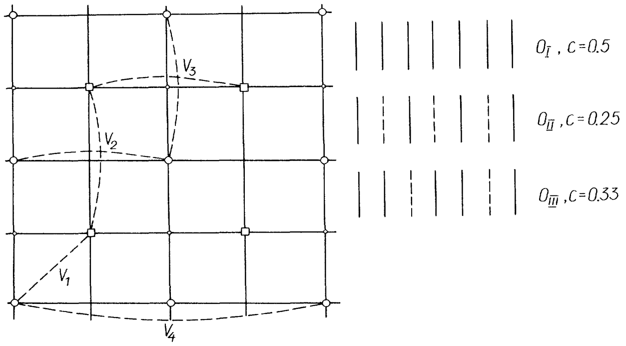

FIG. 1. CuO plane. Filled circles—copper atoms, open circles—oxygen atom sites, squares—vacancies. The ground states of the orthorhombic phases are presented on the right. The solid line represents an occupied chain, the dashed line - a vacancy chain (vacancy sublattice is not shown).

The statistical model which describes the oxygen atoms ordering in 1:2:3 superconductors was suggested by Wille *et al.*[11] In that model the interactions V_1, V_2, V_3 between the pairs of the nearest oxygen atoms in the CuO plane (see Fig. 1) are introduced. The signs of the constants ($V_1>0$, $V_2<0$, $V_3>0$) are chosen to satisfy the conditions of TG, O_I, and O_{II} phase occurrence in $\mathrm{YBa}_2\mathrm{Cu}_3\mathrm{O}_{7-x}$.[3,5] The low-temperature calculation of the (T,c) diagram, which was performed in the lattice-gas model by the modified cluster-field method[12,13] at $V_2/V_1=-0.5$, $V_3/V_1=0.5$ is presented in Ref. 14. In this paper only one-phase regions are found to occur at low T. Narrow mixed-phase regions can appear at higher T and $c<0.25$ (see Fig. 2 in Ref. 14). The estimation of the intra- and inter-O—Cu—O chain correlations has shown that the variation of the oxygen content is related to the changes of the chain segment length. It is worth noting that our results were recently supported by Kikuchi and Choi.[15].

In Sec. II of the present paper the investigation of the (T,c) diagram is carried out in the wide range of the variations of the constants V_1, V_2, V_3. Section III is devoted to the calculation of the diagram when the constant V_2, corresponding to the oxygen interaction via the copper atom, depends on the oxygen content. In this section the problems related with the O_I phase existence at low concentrations are discussed. The changes of the diagram due to the inclusion of next-nearest neighbor interaction are considered in Sec. IV. The calculations are performed in the framework of the model.[12–14]

II. THE VARIATION OF THE CONSTANTS

Here we investigate the changes of the form of the $(t=T/V_1,c)$ diagram due to the variation of the constants V_1, V_2, and V_3. The calculation is carried out by the modified cluster-variation method with the four-point and five-point basic clusters which were also used by Wille *et al.*[11] It should be noted that the variation of the constants in some specific cases is also studied in Refs. 15 and 16.

(1) The O_{II} phase appears in the diagram when the ine-

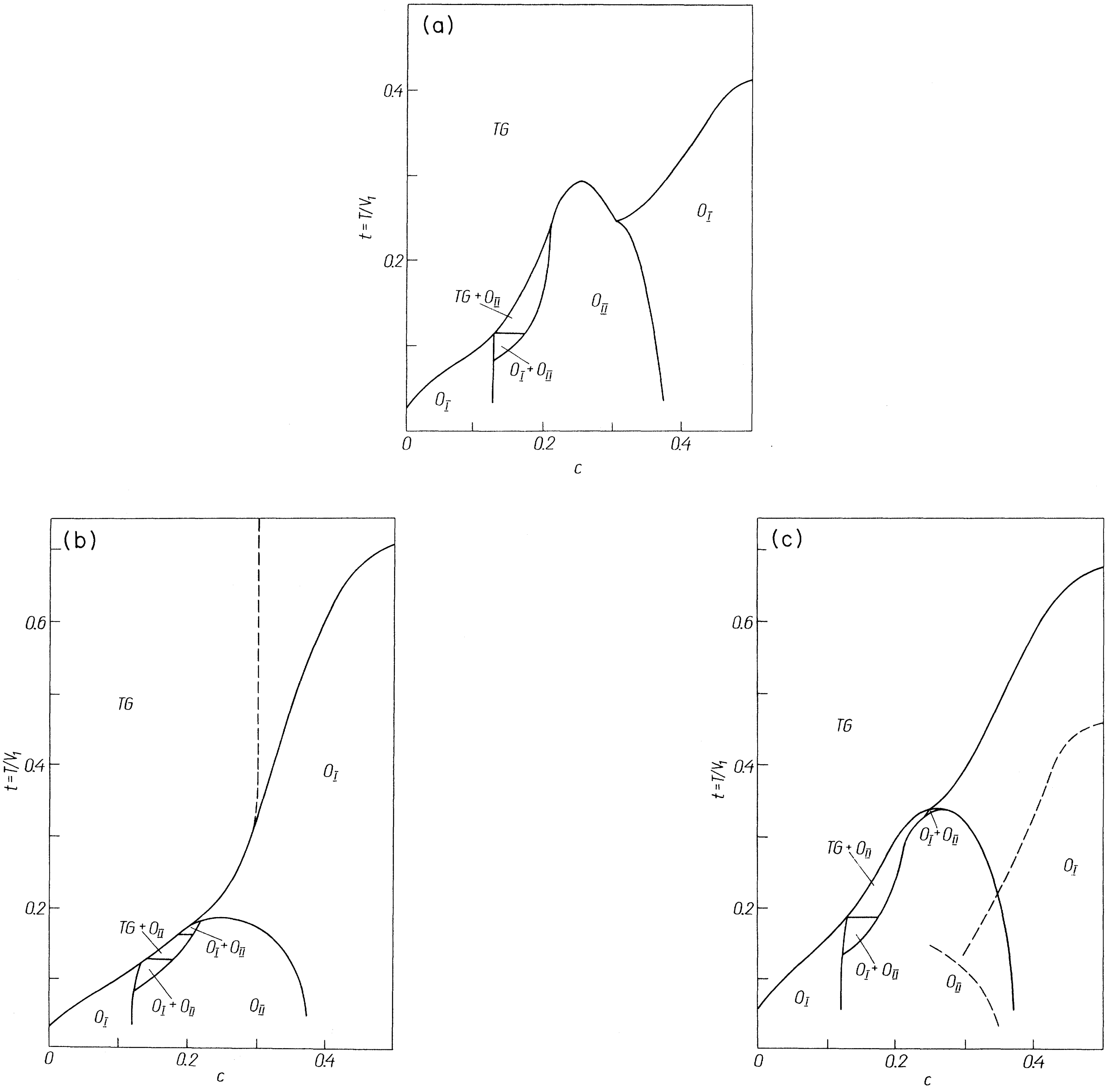

FIG. 2. Phase diagram. (a) $V_2/V_1=-0.5$, $V_3/V_1=0.8$. (b) $V_2/V_1=-0.5$, $V_3/V_1=0.2$ (the dashed line corresponds to the nearest-neighbor exclusion $V_1=\infty$). (c) $V_3/V_1=0.5$, $V_2/V_1=-0.8$ (solid lines) and $V_2/V_1=-0.2$ (dashed lines).

qualities $V_2<0$, $V_1>V_3>0$ are valid. Now, let us fix V_1 and change the V_3 value in the V_3/V_1 ratio. The (t,c) diagrams calculated at $V_2/V_1=-0.5$ and $V_3/V_1=0.8$; 0.2 are presented in Fig. 2 (the diagram at $V_3/V_1=0.5$ was shown in Ref. 14). As is seen from Fig. 2 with the decrease of V_3 the TG-$\mathrm{O_I}$ phase-transition temperatures increase, but the temperatures of the transitions between other phases decrease. The triple-point coordinate c also decreases (e.g., $c_t \approx 0.32$ at $V_3/V_1=0.8$, $c_t \approx 0.27$ at $V_3/V_1=0.5$, and $c_t \approx 0.21$ at $V_3/V_1=0.2$). The decrease of V_3 leads to the widening of the $\mathrm{O_{II}}$ phase region in the vicinity of the triple point. Besides, the additional $\mathrm{O_I}+\mathrm{O_{II}}$ two-phase region occurs at $V_3/V_1=0.2$ and $c \lesssim 0.25$. At $V_3/V_1=0.8$ the interval of concentrations is found in which the transitions between the phases $\mathrm{TG}\rightarrow\mathrm{O_{II}}\rightarrow\mathrm{TG}\rightarrow\mathrm{O_I}$ take place at the constant value of t. It resembles the order-disorder transitions via the intermediate modulated phases. This sequence of transitions is not observed at lower V_3. As is shown the variation of the V_3 constant influences only the upper part of the (t,c) diagram but leaves the low-temperature region $(t \lesssim 0.12)$ unchanged.

(2) The changes of the (t,c) diagram obtained at the variation of $|V_2|$ are qualitatively similar to those in (1). However, some quantitative differences can be observed. In Fig. 2(c) the (t,c) diagram calculated at $V_2/V_1=-0.8$; -0.2, $V_3/V_1=0.5$ is presented. As is seen, with the decrease of $|V_2|$ the values of the transition temperatures decrease.

(3) At $c=0.5$ the half of the CuO plane sites is unoccupied. The chains of vacancies are observed between the occupied O—Cu—O chains. No doubt it is very difficult to locate the additional oxygens into the vacancies and thus to increase the total amount of the oxygen. We know the reports of $c \lesssim 0.6$ (see, e.g., Ref. 17) but we are not sure whether the excess oxygen occupies the vacancies or other possible sites in the neighboring layers. Thus, it looks as if the constant V_1, which describes the repulsion effects of the nearest oxygen and the vacancy sites, ought to be of a considerable value. Otherwise the vacancies would be easily occupied by the additional oxygen atoms. To determine the influence of the constant V_1 on the (T,c) diagram we used the infinite exclusion in the first coordination sphere $V_1=\infty$. Then the oxygen excess is located outside the CuO layer.

The calculation shows that exclusion considerably influences the TG-$\mathrm{O_I}$ transition line. The value of the derivative dt/dc along the line sharply increases compared with the case of the finite V_1. The exclusion caused effect is shown by a dashed line in Fig. 2(b). At low t the form of the diagram remains unchanged.

III. CONCENTRATION DEPENDENCE OF THE INTERACTION PARAMETERS

The dependence of the interaction constants on the concentration of the ordering ions is usually observed when the reconstruction of the system electron spectrum occurs. Metallic alloys[18,19] are the typical representatives of such systems. In the case of 1:2:3 compounds the $V_i(c)$ dependence can be related with a metal-semiconductor transition[20] which causes the reconstruction of the band structure and the electron spectrum of the samples. It should also be noted that the copper atoms may change their valence as the concentration of the oxygen increases.

The most probable dependence of the interaction parameters is assumed to be the $V_2(c)$ since this constant describes the interactions of the oxygens via the copper atoms. In the simplest form it can be expressed as

$$V_2(c)=V_2^0+V_2'c\ , \tag{1}$$

where $V_2^0=V_2(c=0)$. The thermodynamic potential at $V_2'\neq 0$ contains the additional members which are proportional to the values of the short-range correlations of the lattice sites bonded by the V_2 constant. We investigated the possible forms of the (t,c) diagram with $V_2(c)$ from (1) at positive and negative values of V_2^0.

(1) $V_2^0<0$. At $0<V_2'<|V_2^0|/c$ the form of the diagram is qualitatively similar to that at $V_2'=0$. It can be seen from the comparison of Fig. 2 with Fig. 3(a) where the diagram calculated at $V_2'/|V_2^0|=1$ is presented. At $V_2'<0$ the system gains the additional attractive interaction which favors the occurrence of the two phase regions $\mathrm{TG}+\mathrm{O_{II}}$ and $\mathrm{O_I}+\mathrm{O_{II}}$. The area of their existence increases with the increase of $|V_2'|$. In Fig. 3(b) the diagram calculated at $V_2'/|V_2^0|=-1$ is presented. It resembles that conjectured by Wille *et al.*[11]

(2) $V_2^0>0$. At $V_2'>0$ the $\mathrm{O_{II}}$ phase is found to be unstable. This fact disagrees with the most experimental data and can be regarded as unlikely.

(3) V_2 changes the sign. It was stated in the numerous experimental papers (see, e.g., Refs. 2 and 20) that the TG phase is observed at the concentrations $0<c<0.25$. If this data gains support in the future then in order to explain this result by the model described here, we have to make the following assumption. The constant V_2 becomes strongly c dependent at $c=0.25$ since at this point a metal-semiconductor transition is known to occur in the 1:2:3 system [similar behavior of the interaction constants is observed, e.g., in $\mathrm{Ni}_{1-x}\mathrm{Cu}_x$ (Ref. 18)]. Besides, we assume that the constant is positive at $c<0.25$ and negative at $c>0.25$. Then in the linear case (1) $V_2'=-4V_2^0$ and at $c>0.25$ the decomposition of the phases takes place in almost the whole temperature interval. This presumably disagrees with the most experimental facts. Therefore we assume that $V_2(c)$ is weakly c dependent on the left and on the right of the point $c=0.25$ but has almost discontinuity in the vicinity of the point. One of the possible dependences is schematically shown in the inset of Fig. 4. Let us discuss the influence of this $V_2(c)$ dependence on the (t,c) diagram since the concrete calculations of the diagram with such complicated behavior of the $V_2(c)$ constant would be rather premature. When we analyze the lattice-gas model on the square lattice with the repulsive interactions $V_1>0$, $V_2=V_3>0$, we must take into account the result of Binder and Landau[21] of the disordered phase existence in the whole temperature interval at $0<c<0.125$. Besides, the transition line at $0.125<c<0.25$ from the

disordered to the ordered O′ phase with a broken chainlike arrangement is observed at rather low T. If the V_2 constant changes the sign at $c > 0.25$ and becomes negative, the diagram at $0.25 < c < 0.5$ is supposed to be similar to any of the diagrams in Figs. 2 and 3. However, the authors are not sure whether the decomposition of the phases over the wide concentration range does not take place. If not, the diagram corresponding to the $V_2(c)$ dependence from the inset would be like that schematically drawn in Fig. 4.

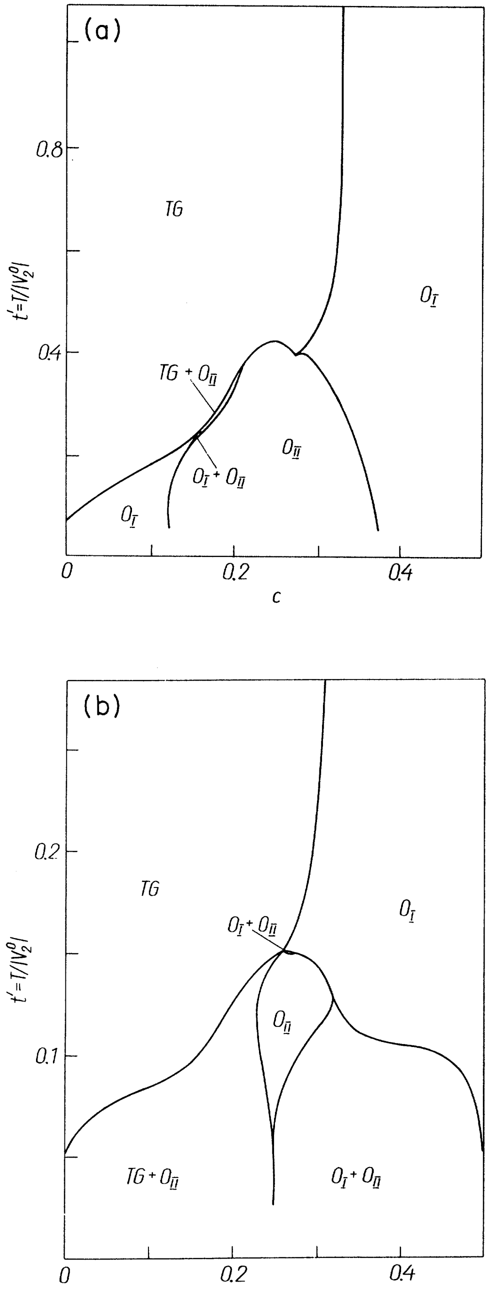

FIG. 3. Phase diagram at $V_2(c)=V_2^0+V_2'c$, $V_3/V_1=1$ and $V_1=\infty$. (a) $V_2'/|V_2^0|=1$. (b) $V_2'/|V_2^0|=-1$.

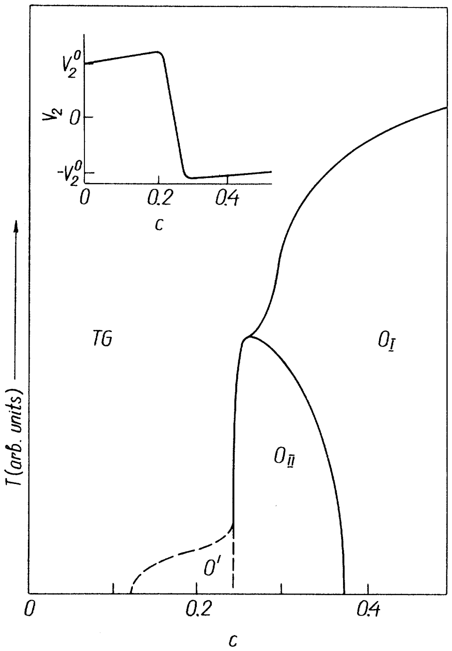

FIG. 4. The proposed form of the phase diagram when the V_2 depends on the concentration in a way shown at the inset. The inset represents almost jumplike $V_2(c)$ dependence which is presumably caused by the metal-semiconductor transition at $c=0.25$.

The assumption of the complicated $V_2(c)$ behavior is not only one which provides the explanation of the TG phase existence at low T and c. It is known[4,22] that the TG phase in $ErBa_2Cu_3O_{7-x}$ is found to exist located at $0<c<0.16$. The location does not depend on the temperature of the measurement [$T=300$ K (Ref. 4) and $T=20$ K (Ref. 22)]. The orthorhombic phase is found at $c>0.16$ in both papers. Thus, the vertical bar of the TG-O_{II} transitions has to appear in the diagram at $c\approx0.16$. This result slightly differs from the assumptions of Fig. 4 at low T and is in marked contrast to the diagrams of Figs. 2 and 3, in which the O_I phase occurs at $O<c<0.125$ and low T. However, one must take into account that a rather small amount of oxygen occupies the 1:2:3 specimen at this interval of the concentrations. Since the establishment of thermodynamic equilibrium in systems with a low concentration of oxygen requires a prolonged time, we do not preclude the possibility of the TG phase to be observed experimentally, though the true thermodynamic phase will be O_I and it will take the corresponding form if one waits a sufficiently long time.

IV. THE NEXT-NEAREST INTERACTIONS

Recently the new orthorhombic superstructures have been found to occur in $YBa_2Cu_3O_{7-x}$ at $c>0.25$.[6,7] The observations have shown that one of these phases O_{III} (see Fig. 1) is thought to be thermodynamically stable, while other phases are presumably metastable and occur due to some external influence. However, it is impossible to obtain the O_{III} phase in the framework of the mod-

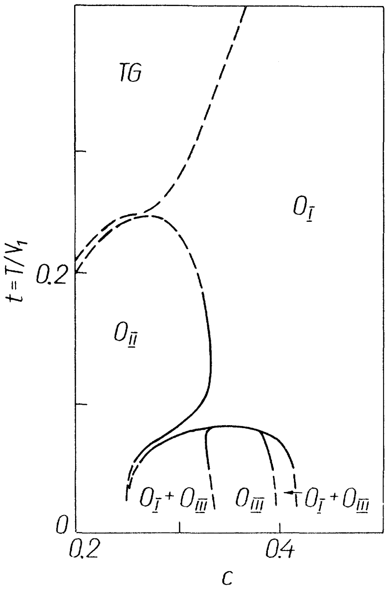

FIG. 5. Phase diagram at $V_2=-0.5$, $V_3=0.5$, $V_4=0.01$. Solid curves represent our calculations, dashed—the data of Ref. 14. The vacancy sublattice and the constant V_1 are not taken into account, although, for convenience, other constants and temperature are normalized to V_1.

el.[11,14] As it was suggested in Refs. 14 and 23 the new phase can become stable if the next-nearest oxygen interactions are considered.

We investigated the changes of the (t,c) diagram when the constant $V_4>0$ was taken into account. In agreement with Ref. 14 the sublattice of the vacancies does not influence the phase diagram since it is almost empty at $t \lesssim 0.2$ and $0.25 \lesssim c \lesssim 0.5$. Therefore the constant V_1 can be neglected in this interval. To make the calculation we have chosen a six-point basic cluster which includes the constants V_2, V_3, V_4.

The obtained (t,c) diagram is plotted in Fig. 5. We added to the diagram the results of the high-temperature calculations[14] since the constant V_4 hardly influences the form of the phase diagram at $V_4<|V_2|, V_3$ and $t>0.2$. As is seen from Fig. 5 the O_I, O_{II}, and O_{III} phases and the O_I+O_{II} two-phase region occur at $0.25 \lesssim c \lesssim 0.5$. With the decrease of temperature the O_{II} phase region decreases and the curve of the second-order phase transition between the O_I and O_{II} phases approaches the $c=0.25$ value. It is worth noting that the direct phase transition between the O_{II} and O_{III} phases is not observed in the diagram. The increase of the constant V_4 leads to the increase of maximum temperature of the O_{III} phase and widens the O_I phase region, confined between the O_{II} phase and the O_I+O_{III} two-phase region.

Our calculation[23] and recent Monte Carlo results[24] have shown that the oxygen arrangement corresponding to the phase with the reduced wave vector $(\frac{2}{5},0,0)$ (Ref. 6) does not occur in the (t,c) diagram at the considered set of the interaction parameters. However, the microislands of this phase can be found interspersed in the O_{III} phase arrangement. It is worth noting that the simultaneous existence in the diagram of the O_{III} phase and the O_I+O_{II} two-phase region turns out to be impossible at $c>0.25$. Really, if the system together with $V_2<0$ has the additional attractive interactions between the O—Cu—O chains (e.g., $V_4<0$) then the decomposition of the phases into the two-phase mixtures at low T necessarily takes place. As a result, the (t,c) diagram would be like that in Fig. 3(b). But when the repulsive next-nearest interactions are included, then the O_{III} phase is found to appear instead of the two-phase region.

V. DISCUSSIONS

The variation of the interaction constants V_1, V_2, V_3 along with the analysis of the V_2 dependence on the oxygen concentration and the consideration of the next-nearest oxygen chain interactions allows us to make the following conclusions.

(1) If we remain within the framework of the interaction model $V_1>V_3>0$, $V_2<0$, the variation of the constants quantitatively changes the (t,c) diagram at high and average temperatures including the triple point. The form of the diagram at low temperatures is very stable and almost independent of the numerical values of the constants. The exclusion in the first coordination sphere makes the value of dt/dc along the tetragonal-orthorhombic transition line almost infinite at high temperatures.

(2) Various phase diagram types can be obtained if the constant V_2 is a linear function of the oxygen amount and $V_2<0$. In the case of additional repulsive interaction $(dV_2/dc>0)$ the diagram is similar to that which is obtained if the V_2 vs c dependence is neglected. The diagram considerably changes at $dV_2/dc<0$ since the additional attraction leads to the decomposition of the system into the $TG+O_{II}$ and O_I+O_{II} two-phase regions at low T.

The region of the tetragonal phase at low c and T can notably increase when V_2, remaining attractive at $c>0.25$, changes the sign at the metal-semiconductor transition point $c=0.25$.

(3) The inclusion of the next-nearest interaction leads to the appearance on the (t,c) diagram of a new orthorhombic superstructure which can be described by the reduced wave vector $(\frac{1}{3},0,0)$. The suggested model causes the decrease of the O_{II} phase region at $c \approx 0.25$. The phase with $(\frac{2}{5},0,0)$ does not occur at the considered set of the interaction parameters.

(4) The analysis carried out allows to estimate the values of the interaction constants of each 1:2:3 superconductor provided the definite experimental data are known. Certainly, the precise values of the constants still cannot be unambiguously found, but qualitative estimations of V_1, V_2, and V_3 can be performed just now on the basis of the TG-O_I phase-transition line data and the triple-point coordinate.

In agreement with the data[2] the triple point of $YBa_2Cu_3O_{7-x}$ is found at $c_t \approx 0.31$, $T_t \approx 740$ K. At $V_2/V_1=-0.2$, $V_3/V_1=0.5$ we obtain the concentration of the triple point $c_t \approx 0.29$ which is rather vicinous to experimental. The value of the triple-point temperature t at the given set of the interaction parameters is equal to 0.12. Then $V_1 = T_t / t_t = 0.54$ eV and correspondingly $V_2 = -0.11$ eV, $V_3 = 0.27$ eV. Besides, the derivative of the TG-O_I phase transition temperature with respect to the oxygen concentration dT/dx is equal to $V_1 dt/dx$. In the vicinity of the triple point $dt/dx = 0.45$ at $V_2/V_1 = -0.2$, $V_3/V_1 = 0.5$. As a result, we obtain $dT/dx = 2.8 \times 10^3$ K which is found between the experimental data $dT/dx = 2.2 \times 10^3$ K (Ref. 2) and $dT/dx = 4.5 \times 10^3$ K.[4]

The values of the constants obtained here differ considerably from the estimations $V_1 = 0.09$ eV, $V_2 = -0.2$ eV, $V_3 = 0.05$ eV (Ref. 15), $V_1 = 0.12$ eV, $V_2 = -0.04$ eV, $V_3 = 0.017$ eV (Ref. 25), and $V_1 = 0.192$ eV, $V_2 = 0.144$ eV, $V_3 = 0.096$ eV.[16] These values were obtained by matching them with the experimental line of the TG-O_I transition.[3,26] However, the shift of dT/dx vicinous to the experimental can be obtained with various sets of V_1, V_2, V_3. In order to make the comparison less ambiguous one must choose a certain starting point, e.g., the triple point as in our case. The dependence of this point coordinate on the V_1, V_2, V_3 variation is more pronounced than that of the dT/dx shift. Certainly, it should be noted that the determination of the triple-point coordinate is connected with considerable experimental complications which arise due to the instability of the system in the vicinity of the triple point.[2]

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