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The Effect of Pressure on the Crystal Structure and Critical Temperature of the Superconductor YBCO

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ABSTRACT

The effect of pressure on the crystal structure and the transition temperature of the high temperature superconductor $YBa_2Cu_3O_{7-x}$ (x = 1, 0.5 and 0) was studied. Main force of interaction is the electrical interaction because of the ionic nature of these compounds, model of the crystal structure based on experimental data was established. The pressure effect on the above structure was considered, also considered details of ionic position, inter - ionic distances and forces between them. Madelung method used for calculating the forces between the ions and their binding energy. Because Madelung constant reflects the real crystal structure. The effect of pressure on YBCO structure was found via calculating the bulk modulus. The relative volume of YBCO decreases with increasing pressure and oxygen content in the compound. elongation of the Cu(2)-O(4) bond was detected under pressure along the b direction also a change of Madelung energy was found which gives an indication of the effect of pressure on T_c .

الملخص

في هذا البحث تم دراسة تأثير الضغط على التركيب البلوري ودرجة حرارة التحول للمركب الفائق التوصيل ذو الدرجة الحرارية العالية ($YBa_2Cu_3O_{7-x}$ (x = 1, 0.5 , 0. القوة الكهربائية تمثل القـوة الأساسية لأن المركب أيوني. نموذج التركيب البلوري بني على أساس بيانات تجريبية. ولغرض دراسـة تأثير الضغط على هذا المركب مراعين التفاصيل الدقيقة للمواقع الأيونية وأطوال الأواصر والقـوى بـين الأيونات. تم استخدام طريقة مادلونج لحساب القوى بين الأيونات وطاقة الربط بينها لأن ثابت مـادلونج يعكس التركيب البلوري فعليا. كما أن تأثير الضغط على المركب حسب من خلال معامل الصلادة. ولقـد وجد أن الحجم النسبي للمركب YBCO، يقل بزيادة الضغط ونقصان نسبة الأكسجين في المركب. ولقـد وجدنا زيادة في طول الأصرة Cu(2)-O(4) بفعل الضغط باتجاه الإحداثي b ووجد هناك تغير في طاقة مادلونج مما يشير إلى تأثير الضغط في درجة حرارة تحول المركب.

INTRODUCTION

Since the discovery of the high temperature superconductivity HTSC a great efforts have been done to increase the transition temperature T_c (Chu, 2002), this challenge, lead

to thorough experimental and theoretical work. The pressure effect was one of the ways to tackle this problem (Gensbers et al., 1999). The pressure have many effects on the superconductor. In a study done by (Huber and Liverman, 1990) found a pressure dependence for $YBa_2Cu_3O_7$. On the other hand (Hiroki, 1989) found a change in the bond length Cu-O under the effect of pressure, (Sadewasser et al., 1999), have studied the dependence of T_c on the hydrostatic pressure in $YBa_2Cu_3O_7$ also the same group (Sadewasser et al., 2000), proceed more, on the same direction. It was proved by many workers that T_c is a function of pressure and is increased with increase of pressure (Rabinowitz, 1997), especially in hole doped superconductors.

(Enquiraiz, 2001) studied the change in the bond length under pressure (Khosroabadi et al., 2002), found a change in the ionic positions, in the bond length, and the distances between the planes of Cu-O. (Klatz and Schilling) had noticed that little number of theorations have tried to study this problem (Rabinowitz and Macmillan, 1997).

The aim of this study is to study the effect of pressure on the crystal structure of $YBa_2Cu_3O_{7-\delta}$ and its effects on T_c, considering the superconductor, as an ionic compound. Then the compressibility was determined, so the relation between the pressure and the crystal parameters was found.

THEORY

To formulate the theoretical basis for this study. The crystal structure of $YBa_2Cu_3O_x$ should considered through the positions and the distances between the atoms forming the crystal. On the basis of this crystal model Madelung constant determined, then the bulk modulus of YBCO found, and the pressure effect on this crystal can be found as well.

The Crystal Structure of $YBa_2Cu_3O_7$:

The crystal of YBCO is composed of three units with pervoskite structure. The copper ions are on the apices of the pervoskite cubes, Y in the center of the crystal, Ba ions in the centers of the upperand lower cubes and O ions on the middle of the edges as shown in fig. (1)

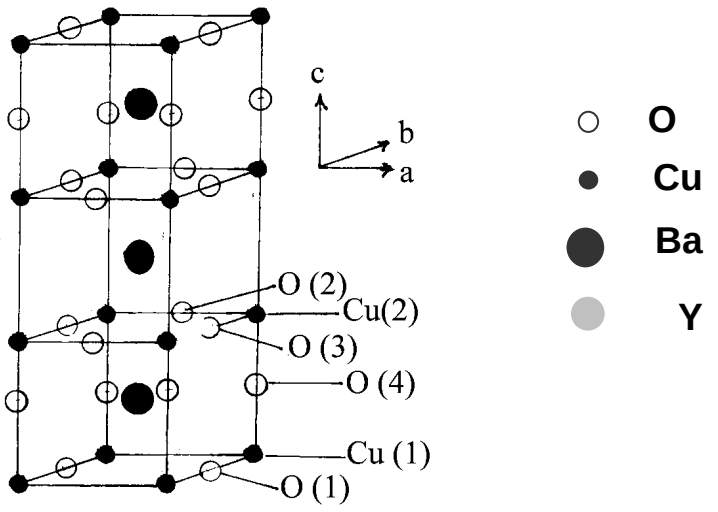

Figure 1 : The crystal structure of $YBa_2Cu_3O_7$. (Sharp, 1990)

One of the lost O atoms are from the O(4) positions around the Y atom. The other atom are lost from the O(1) positions in the upper and lower cubes. This lead to the formations of a pyramids in the upper and middle cubes with Cu-O basis and a lamella of Cu-O in the lower cube as shown in fig. (2).

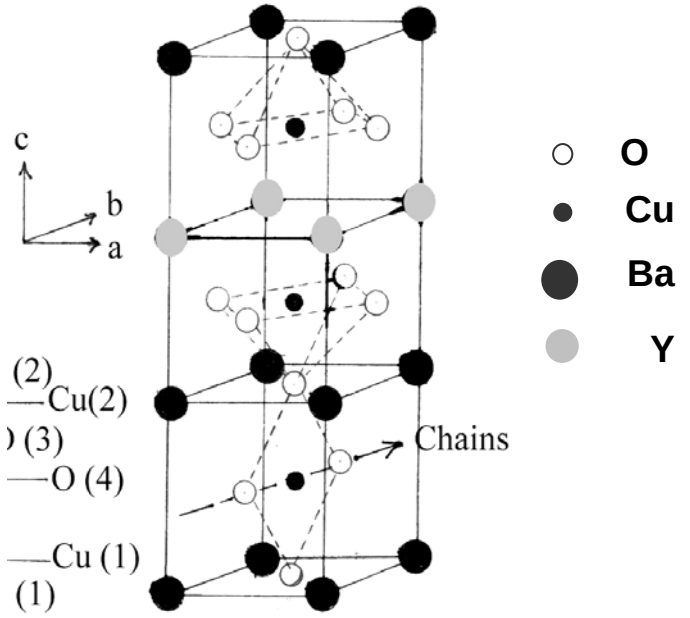

Figure 2: Another representation of the $YBa_2Cu_3O_7$ crystal structure showing the plains and the chains of Cu-O. (Sharp, 1990).

It was shown by many studies that there is a great role for these chains (Hiraoka, 1989) We will study the compound YBCO for different v O content values, in the figure (3), these structures are shown:

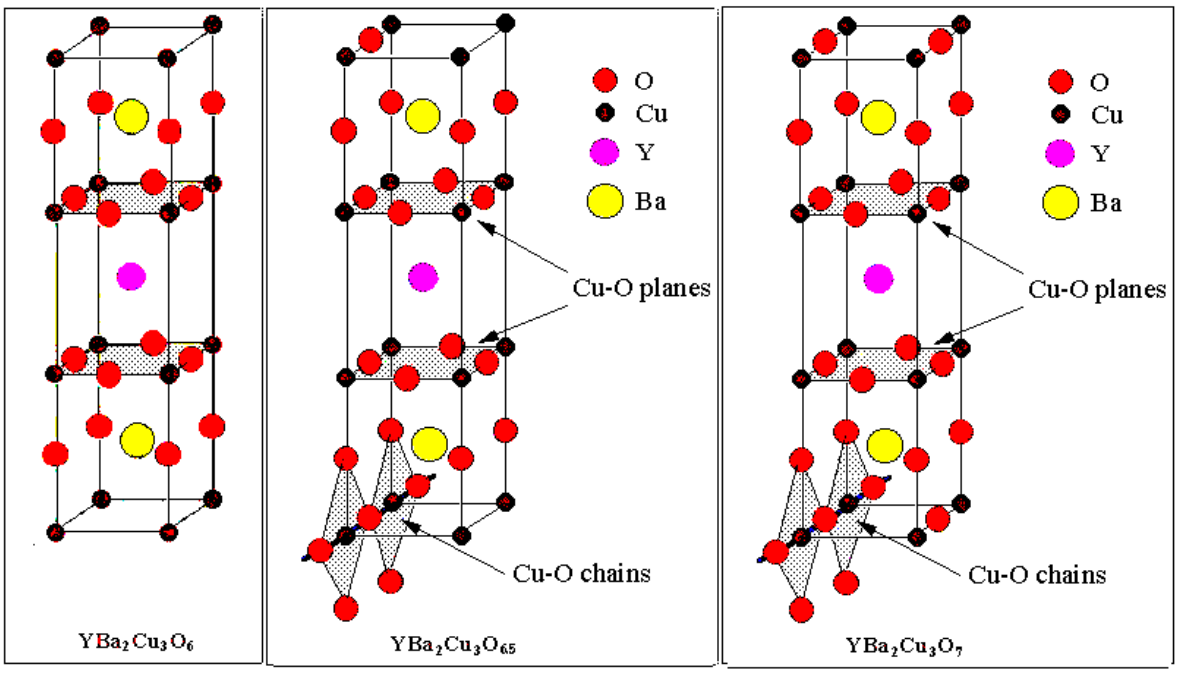

Figure 3: The Cu-O chains and plains in the compounds $YBa_2Cu_3O_7$, $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_6$. (Cyrot and Pavuna, 2004)

Electrical Aspects of YBCO:

To calculate the compressibility we have to calculate the Madelung energy. To do this we must have an idea about the crystal structure and positions of the ions and their respective charges.

The interaction energy between the ion i and all other ions j is Ui this is equal:

$$U_i = \sum_{j \neq i} U_{ij} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

The interaction energy term U_{ij} involve, electrical attraction and repulsion. Another Source of interaction comes from the overlap of orbitals of interacting ions and has repulsive character. So the interaction term can be written as:

$$U_{ij} = \lambda \exp(-r_{ij} / \rho) \pm \frac{q^2}{r_{ij}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

Let the interaction distance $r_{ij} = P_{ij} R$, where P_{ij} represent a geometrical factor depends on the crystal under study, R represent the nearest neighbor distance, λ empirical parameter account for the repulsive interaction due to the overlap of orbitals. The total interaction energy for N molecules in the crystal is:

$$U_{tot} = NU_i \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

By connecting the above equations we have:

$$U_{tot} = NU_i = N\left(z\lambda e^{-R/\rho} - \frac{\alpha q^2}{R} \right) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4a)$$

$\alpha = \sum_j \frac{(\pm)}{P_{ij}}$ is called the Madelung constant. At the equilibrium state $\frac{dU_{tot}}{dR} = 0$;

$$N \frac{dU_i}{dR} = -\frac{Nz\lambda}{\rho} \exp(-R/\rho) + \frac{N\alpha q^2}{R^2} = 0 \quad ; \text{or}$$

$$R_0^{\,2} \exp(-R_0 / \rho) = \frac{\rho \alpha q^2}{z\lambda} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4b)$$

Substitution of λ from equation (4b) in (4a) lead to:

$$U_{tot} = -\frac{N\alpha q^2}{R_0}\left(1 - \frac{\rho}{R_0}\right) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (5)$$

Is equal to at nearest neighbor distance R_o: The quantity $-\frac{N\alpha q^2}{R_0}$ is called Madelung Energy, which determines the cohesive energy. The above relation is very important for us, because we can find the effect of pressure on the crystal via the bulk modulus, as follows:

$$P = -\frac{dU}{dV} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (6)$$

$$\frac{dp}{dV} = -\frac{d^2U}{dV^2} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (7)$$

For the YBCO it consist from three pervoskite cubes of volume $a=\sqrt{2}R$ therefor, volume of N molecules is :

$$V = N\left(\sqrt{2}\right)^3 R^3 \quad \text{(8)}$$

$$\frac{dR}{dV} = \frac{1}{dV/dR} = \frac{1}{3N\left(\sqrt{2}\right)^3 R^2} \quad \text{(9)}$$

$$\frac{d^2U}{dV^2} = \frac{d^2U}{dR^2}\left(\frac{dR}{dV}\right)^2 + \frac{dU}{dR}\frac{d^2R}{dV^2} \quad \text{(10)}$$

At equilibrium $\frac{dU}{dR}=0$, at $R=R_0$, therefor the Bulk modulus B is

$$B = V\frac{d^2U}{dR^2}\left(\frac{1}{3N(\sqrt{2})^3 R^2{}_0}\right)^2 \quad \text{(11)}$$

By taking the second derivative from equation (5) we have

$$B = \frac{\alpha q^2}{18\sqrt{2}R^4{}_0}\left(\frac{R_0}{\rho}-2\right) \quad \text{(12)}$$

The value of ρ was determined from experimental of bulk modulus (Khosroabadi et al., 2004). The bulk modulus can be written as $B=-V\frac{dp}{dV}$ normally the value of the bulk modulus can be found from measurements of the volume compressibility which can expressed as ($\kappa = \frac{1}{B}$) (Rahinowitz and Macmillan, 1997).

Then the volume compressibility can expressed as:

$$\kappa = -\frac{1}{V}\frac{dV}{dP} \quad \text{(13)}$$

$$\int_{p_1}^{p_2} \kappa dP = \int_{V_o}^{Vp} -\frac{1}{V}dV \quad \text{(14)}$$

$$K\Delta P = -\ln\frac{V_P}{V_O} \quad \text{(15)}$$

Where $\Delta P = P_2 - P_1$;

$$e^{-K\Delta P} = \frac{V_P}{V_O} \quad \text{(16)}$$

$$V_P = V_0 e^{-K\Delta P} \quad \text{(17)}$$

From the former equations we can find the change in the volume of the unit cell for YBCO as a function of pressure:

$$= P = \frac{B_0}{A}\left[\left\{\exp A\left(1-\frac{V_p}{V_0}\right)\right\}-1\right] \quad \text{(18)}$$

$$= A = \frac{n+10}{3}$$

The parameter A is equal to 6.16 for ($YBa_2Cu_3O_{6.5}$) and 6.15 for ($YBa_2Cu_3O_7$) (Rabinowitz and Macmillan, 1997), (Kumar, 1997).

Shift In Ionic Distances:

To study the shift in the ionic distances we return to equation 5 and count (Madelung Energy) about certain ionic, like O(4), O(3), Cu(2),…. etc. For this we redraw fig. (1), by making half diagonal transitions as in fig. (4).

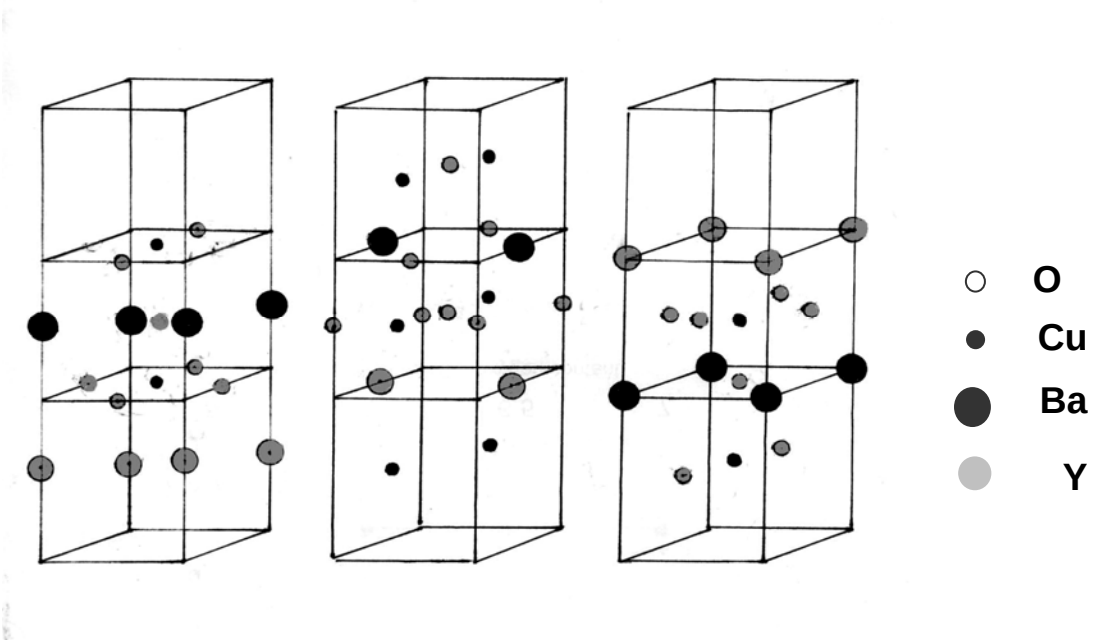

Figure 4 : YBCO crystal structure after transitions.

Madelung Energy can be calculated as follows:

Madelung Energy = $W_c = \frac{1}{2}\sum_{i}^{cell}\sum_{j,j\neq i}\frac{q_i q_j}{R_{ij}} = \frac{1}{2}\sum_{i}^{cell} q_i \varphi_i$ ……..(19)

This equation is written for certain cell i, in which the summation goes over all ions j of the cell except that when j = j. Then φ_i represent Madelung potential at the ion i.

The data for the ionic positions and dimensions for the crystals of YBCO compounds are shown in Tables (1, 2 and 3).

Table 1 : The dimensions (a, b and c) of $YBa_2Cu_3O_6$, $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_7$. (Wright and Balter, 1990)

Lattice parameters (A^0)	**$YBa_2Cu_3O_6$**	**$YBa_2Cu_3O_{6.5}$**	**$YBa_2Cu_3O_7$**
a	3.8630	3.8430	3.8231
b	3.8630	3.8746	3.8863
c	11.8300	11.7554	11.6809

Table 2 : The ionic positions (X, Y and Z) for $YBa_2Cu_3O_6$, $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_7$. (Wright and Balter, 1990)

Ion	**Atomic position(X,Y,Z) (A^0)**		
	$YBa_2Cu_3O_6$	**$YBa_2Cu_3O_{6.5}$**	**$YBa_2Cu_3O_7$**
Y	(0.5 ,0.5 ,0.5)	(0.5 ,0.5 ,0.5)	(0.5 ,0.5 ,0.5)
Ba	(0.1934 ,0.5 ,0.5)	(0.1888 ,0.5 ,0.5)	(0.1843 ,0.5 ,0.5)

Table 3 : The distances between ions in $YBa_2Cu_3O_6$, $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_7$. (Wright and Balter, 1990)

Dimensions	**Inter-ionic distances(A^0)**		
	$YBa_2Cu_3O_6$	**$YBa_2Cu_3O_{6.5}$**	**$YBa_2Cu_3O_7$**
Cu(1)-O(1)	1.931	1.937	1.943
Cu(1)-O(4)	1.814	1.832	1.850
Cu(2)-O(2)	1.944	1.936	1.928
Cu(2)-O(3)	1.944	1.953	1.962
Cu(2)- O(4)	2.450	2.377	2.304
Y-O(2)	2.402	2.408	2.415
Y-O(3)	2.402	2.390	2.378
O(1)-O(4)		2.666	2.683
O(2)-O(3)	2.732	2.729	2.726
2O(2)- O(2)	2.856	2.861	2.867
O(3)-O(3)	2.856	2.842	2.829
Ba-O(1)		2.879	2.879
Ba-O(2)	2.927	2.951	2.976
Ba-O(3)	2.927	2.948	2.970
Ba-O(4)	2.772	2.760	2.743
Cu(2)-Cu(2)	3.303	3.338	3.373
Cu(1)-Cu(2)	4.264	4.209	4.154

Table 4: The ionic shift for some of the ions in $YBa_2Cu_3O_7$In Z direction by theapplication of pressure in the directions a and b the negative sign indicating moving toward Cu-O chains (+) sign means the reverse. (Pickett, 1997)

Ion	**The shift in the Z direction by the application of 2.5 Gap in the a direction**	**Shift in the Z-direction for a pressure 2.5 Gpa on the direction b**
O(2)	0.023-	0.033-
O(3)	0.005-	0.008+
O(4)	0.003-	0.018+
Cu(2)	0.015-	0.013+
Ba	0.006-	0.008+

RESULTS AND DISCUSSION

In this research we apply the results of the theoretical calculation about the effect of pressure on the structure of the superconductor $YBa_2Cu_3O_{7-\delta}$ ($\delta = 0, or 0.5, or 1$), we calculate then the change in volume after the calculation of the compressibility on the basis of the distances and charges of ions then finding the Madelung factor for each of these structure. These calculations are lengthy and the results of this effort is shown in figure (5), where the relative volume of the superconductor $YBa_2Cu_3O_{7-\delta}$ was drawn as a function of pressure for ($\delta = 0, or 0.5, or 1$). The figure shows clearly that : the lower the oxygen content, the greater the ability for compression for pressure 10 GaP there is decrease about 5.1% this very near to the result of (Khosroabadi et al., 2004), which was (5.7%). In order to understand the effect of pressure on the structure and alternatively on the superconductivity a study of the shift of the ionic position was done the results are shown in table 5, 6 and 7 and compared with the Table (4) and for the inter-atomic distances comparison done with Tables (2 and 3).

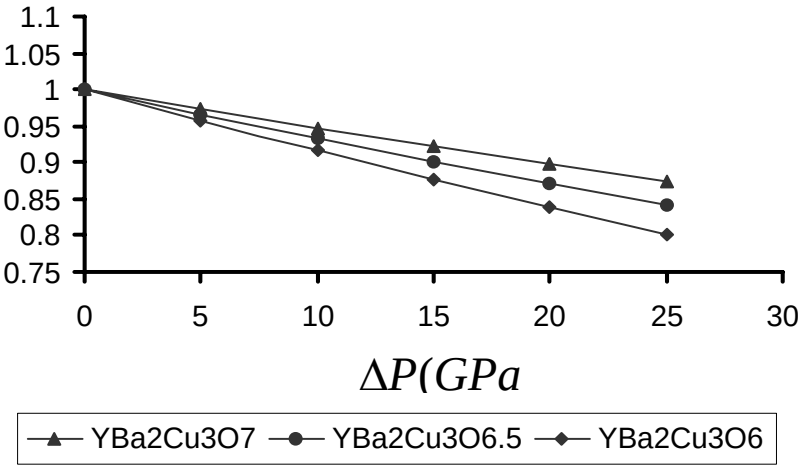

Figure 5 : The relative change in unit cell of YBCO as a function of pressure for different values of oxygen content.

Table 5: The inter-ionic distances and Madelung potential (M.P) for $YBa_2Cu_3O_7$ under pressure along a and b directions on the site O(4).

P(Gpa)	Inter-ionic Distances(A^0)							.P.at site O(4) (eV)
	O(4)-Ba	O(4)-Y	O(4)-Cu(1)	O(4)-Cu(2)	O(4)-O(1)	O(4)-O(2)	O(4)-O(3)	
0	2.743	4.832	1.850	2.304	2.683	3.004	3.026	-20.824
2.5 (a)	2.743	4.835	1.847	2.292	2.680	2.980	3.015	-20.803
2.5(b)	2.743	4.817	1.868	2.299	2.695	2.989	3.030	-20.787

Table 6 : The inter-ionic distances for and the M.P. for $YBa_2Cu_3O_7$ along a and b directions on the site O(3).

P(Gap)	Inter-ionic Distances(A^0)								M.P.at site O (3) (eV)
	O(3)-Ba	O(3)-Y	O(3)-Cu(1)	O(3)-Cu(2)	O(3)-Cu(2)*	O(3)-O(1)	O(3)-O(2)	O(3)-O(4)	
0	2.970	2.378	4.594	1.962	3.902	4.154	2.726	3.026	-19.400
2.5 (a)	2.968	2.381	4.576	1.962	3.932	4.134	2.726	3.015	-19.315
2.5 (b)	2.970	2.371	4.613	1.962	3.873	4.175	2.726	3.030	-19.489

Table 7 : The inter-ionic distances for and the M.P. for $YBa_2Cu_3O_7$ along a and b directions on the site Cu(2).

P(Gpa)	Inter-ionic Distances(A^0)							M.P.at siteCu(2) (eV)
	Cu(2)-O(1)	Cu(2)-O(2)	Cu(2)-O(3)	Cu(2)-O(4)	Cu(2)-Ba	Cu(2)-Y	Cu(2)-Cu(1)	
0	4.586	1.928	1.962	2.304	3.318	3.205	4.154	-24.210
2.5 (a)	4.572	1.928	1.962	2.292	3.312	3.213	4.139	-24.237
2.5 (b)	4.598	1.928	1.962	2.299	3.320	3.198	4.167	-24.223

Also calculated the change in M.P. on the application of pressure along the a and b directions as in Table (8).

Table 8: M.P. and the Change in it under the application of pressure along the a and b directions.

Ion	V_{Zero}(eV)	ΔV_a(eV)	ΔV_b(eV)
O(4)	20.824-	0.021	0.037
O(3)	19.400-	0.085	0.089-
Cu(2)	24.210-	0.027-	0.013-

Table 9: Calculated values of some important inter-ionic distances under 0 pressure and 2.5 Gpa on the directions a and b .

Inter-ionic Distances(A^0)	P(Gpa)		
	0	2.5 in the a direction	2.5 in the b direction
Cu(1)-Cu(2)	4.154	4.139	4.167
Cu(2)-Cu(2)	3.373	3.403	3.347
Cu(1)-O(4)	1.850	1.847	1.868
Cu(2)-O(4)	2.304	2.292	2.299

The study of the bond - lengths between the atoms constituting YBCO which have certain nomination as in Figure (2), is of essential importance. For example the bond Cu(1)-Cu(2) inform about the elongation of the upper and lower cubes in the $YBa_2Cu_3O_7$ compound. On the other hand the Cu(2)-Cu(2), points to the elongation of the middle cube, the Cu(1)-Cu(4), Cu(2)-Cu(4) have special importance because O(4) ion represent a chain between the two bonds (Khosroabadi et al., 2002).

In this research the effect of pressure on the bond lengths was studied and the results are shown in tables 5, 6, 7 and 9, the values of the bond lengths can be well compared with the experimental results in Table (4): The distances in O(4)-Cu(2) and O(4)-Cu(1) decreases (about 0.003 A^o) under pressure in the direction a from its value under zero pressure this coincides with (Khosroabadi et al., 2004). The distance O(4)-Cu(2) decrease (about 0.004 A^o) whereas the distances O(4)-Cu(1) increases (about 0.016 A^o) at pressure in the direction b this confirms works (Khosroabadi et al., 2002). Also the distances Y-O(4) increases under pressure (about 0.004 A^o) in the direction a from its value under zero pressure, whereas the reverse occur for the b direction (increase about

0.012 A^o). Proceeding calculations on the same way for all the bonds in the crystal of YBCO the results are as follows:

pressure in the a direction:

Normal distances	**Increasing distances**	**Decreasing distances**
O(4)-Ba	O(4)-Y	O(4)-Cu(1)
O(3)-Cu(2)	O(3)-Y	O(4)-Cu(2)
O(3)-O(2)	O(3)-Cu(2)*	O(4)-O(3)
Cu(2)-O(2)	Cu(2)-Y	O(4)-O(1)
Cu(2)-O(3)	Cu(2)-Cu(2)	O(4)-O(2)
		O(3)-Cu(1)
		O(3)-O(1)
		O(3)-O(4)
		Cu(2)-O(1)
		Cu(2)-Ba
		Cu(2)-Cu(1)
		Cu(2)-O(4)

Pressure in the b direction:

Normal distances	**Increasing distances**	**Decreasing distances**
O(4)-Ba	O(4)-Cu(1)	O(4)-Cu(2)
O(3)-Cu(2)	O(4)-O(3)	O(4)-Y
O(3)-O(2)	O(4)-O(1)	O(4)-O(2)
Cu(2)-O(2)	O(3)-Cu(1)	O(3)-Cu(2)
Cu(2)-O(3)	O(3)-O(1)	O(3)-Y
	O(3)-O(4)	Cu(2)-Y
	Cu(2)-O(1)	Cu(2)-O(4)
	Cu(2)-Ba	Cu(2)-Cu(2)
	Cu(2)-Cu(1)	

The study of (Khosroabadi et al., 2002) introduce a relation between T_c and the bond length of Cu(1)-O(4). Calculations of this study using the ionic model indicate such an increase in this bond. On the other hand the pressure along a direction decreases the bond. This means that pressure increases the transition temperature.

On the other hand counting the change in M.P. around the ions O(4) and Cu(2) under pressures using the results o f table 8 along a and b shows appreciable difference as follows :

$$\Delta V_a^{O(4)} - \Delta V_a^{Cu(2)} = 0.048....eV \quad , \quad \Delta V_b^{O(4)} - \Delta V_b^{Cu(2)} = 0.050....eV$$

This means elongation along b and compression along a under the effect of pressure This gives another evidence that pressure increases the transition temperature.

CONCLUSION

1–The application of the ionic model to study the effect of pressure on YBCO proved to be a successful one . This model consider the main force in the YBCO is the electrostatic attraction and repulsion in addition to Pauli overlap of orbit repulsion .

2–This model was applied using Madelung method to count the interaction force (electrical force (attraction or repulsion), overlap of orbitals repulsion) between a reference ion and other ions in the crystal, then bulk modulus was found, and thus a relation was established between the pressure and the crystal structure .

3–By applying the above procedure ,it was found that the decrease in the relative volume increase with decreasing oxygen content in YBCO, this confirm the experimental result about the effect of pressure on YBCO. Also confirms the role of oxygen in superconductivity .

4–The study calculate the distances between ions, lengths of ionic bonds, distances between Cu-O planes. The study confirms the experimental results about elongation of the bond O(4)-Cu(2) under pressure which is also increases with T_c .

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