Iraqi Journal of Physics, 2008 *Vol. 5, No. 1, PP 75-80*

Effect of Pressure on the Properties of $HgBa_2Ca_2Cu_3O_{8+\delta}$ HTSC System

G. Y. Hermiz

Department of Physics, College of Science, University of Baghdad

Abstract:

High temperature superconductors with a nominal composition $HgBa_2Ca_2Cu_3O_{8+\delta}$ for different values of pressure (0.2,0.3, 0.5, 0.6, 0.9, 1.0 & 1.1)GPa were prepared by a solid state reaction method. It has been found that the samples were semiconductor P=0.2GPa.while the behavior of the other samples are superconductor in the rang (80-300) K. Also the transition temperature T_c=143K is the maximum at P is equal to 0.5GPa. X-ray diffraction showed a tetragonal structure with the decreasing of the lattice constant c with the increasing of the pressure. Also we found an increasing of the density with the pressure.

الفائق التوصيل $HgBa_2Ca_2Cu_3O_{8+\delta}$ تأثير الضغط على خواص لنظام

غزالة يلدا هرمز

قسم الفيزياء / كلية العلوم /جامعة بغداد

الخلاصة

حُضرّ المركب $HgBa_2Ca_2Cu_3O_{8+\delta}$ الفائق التوصيل وبضغوط مختلفة (0.2,0.3,0.5,0.6,0.9,1.0&1.1)GPa بطريقة تفاعل الحالة الصلبة.لقد وجد أن العينات المحضرة وبضغط P=0.2GPa. تظهر خواص شبه موصل بينما بقية النماذج تكون فائقة التوصيل ضمن درجات الحرارة المستخدمة (K 80-300). كما لوحظ أن أعلى درجة حرارة حرجة T_c=143K وعند ضغط 0.5 GPa. بينّت تحليلات الأشعة السينية أن المركب ذو تركيب رباعي tetragonal وأظهرت هذه التحليلات تناقص في قيمة ثابت الشبيكة c مع زيادة الضغط. كما ويلاحظ زيادة في قيمة الكثافة مع زيادة الضغط.

Introduction

The most interesting series of all high temperature cuprate superconductors are $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ in comparable with other families discovered previously. The first reason for this is the high critical transition temperature, the second one is the anisotropy of these HTSC phases is in between Y and Bi bearing cuprates and they are therefore, expected to show effective flux pinning and microstructure exhibiting good texturing of grains [1].

The system Hg-Ba-Ca-Cu-O contains three superconducting phases which can be characterized by a general formula $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$. The synthesis of phase pure samples of these materials has been found very complicated due to high volatility of mercury at elevated temperatures when the phase formation occurs. The first member of the family (n=1), has a critical temperature of 94K [2]. The second one (n=2), has T_c =127K[3]. The third member of this family (n=3) has a sharp superconducting transition at 133K [4].

Application of pressure can increase the critical temperature of the second member to 154K and that of the third to 160K [5]. The addition of isovalent elements with respect to Hg gives a dramatic expansion of the mercury-based family. It is worth mentioning the addition of Tl and Sr will create a new phase, $Hg_{1-x}Tl_xBa_2(Ca_{1-y}Sr_y)_2Cu_nO_{2n+2+\delta}$ and exhibited a critical temperatures of about 132K with y=0.14 and x=0.25-0.5 [6].

Wu *et al* [7] reported that substitution of Pb for Hg in the $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Hg-1223,Tc~135 K) results in an increase of superconducting transition temperature to T_c=143K (but zero-resistance T_c=135K) for samples with nominal composition of $Hg_{0.7}Pb_{0.3}Ba_2Ca_2Cu_3O_{8+\delta}$. The lattice parameter changes resulting from the Pb substitution are too small to explain the T_c change as arising from an effective pressure change.

Gonzalez *et al* [8] studied the phase stability under pressure in the range (0.1MPa-5GPa) for $HgBa_2CuO_{4+\delta}$ (Hg-1201) superconducting system and also measure its compressibility and bulk modulus. They found that the compound exhibited anisotropy elastic properties. The axial compressibility along the c axis was measured to be $(3.96 \pm 0.35)\times10^{-3}GPa^{-1}$ and along the axis $(3.42\pm0.13)\times10^{-3}GPa^{-1}$, correspondding to an anisotropy ratio of 1.16±0.11. The bulk modulus was determined to be (94.7±4.2)GPa and, assuming a Poisson's ratio of 0.2, Young's modulus was estimated to be (170±8)GPa.

Antipov *et al* [9] analyzed the crystal structure of the superconductor $HgBa_2CaCu_2O_{6+\delta}$ by neutron diffraction techniques at room temperature and at 10K. They found that the compound has lattice parameters a=3.8526, c=12.6367Å and T_c =104 K. Annealing in O_2 (300°C for 2 h) reduces the oxygen content to 0.28 Bougrine *et al* [10] report the temperature dependence of the resistivity, thermopower and thermal conductivity measurements of HgBaCaCuO (1223) polycrystalline superconductors, with nominal composition $Hg_{1.4}Ba_2Ca_2Cu_3O_y$.

In this paper we studied the effect of pressure on the oxygen content, transition temperature and the structural properties of $HgBa_2Ca_2Cu_3O_{8+\delta}$ HTSC system.

Experimental

The samples were prepared by solid-state reaction. Appropriate amounts of the powder materials $CaCO_3$, CuO and $BaCO_3$ were mixed together, the mixture were grounded and reground many times to produce a fine powder and then calcined in air at 800°C for 20h in two stages. The calcined powder was regrind again after the mixing of HgO with it and pressed into disc-shaped pellets with different pressure (0.2, 0.3, 0.5, 0.6, 0.9, 1.0 and 1.1)GPa. The pellets were sintered in air at 860°C for 200h with a rate of 60°C/h in a tube furnace and then cooled to room temperature by the same rate of heating. The samples were examined by Meissner effect to evaluate the superconducting state. Iodometric titration was used to find the oxygen content (δ) in the samples. Four probe dc method at temperature range (77-300)K was used to measure the resistivity (ρ) and to determine the critical temperature (T_c)

The structure of the prepared samples was obtained by using x-ray diffractometer (XRD) type (Philips) with the Cu_{Ka} source. A computer program has been used to calculate the lattice parameters, which is based on Cohen's least square method. The lattice parameter for the unit cell could be found from the relation:

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}$$

where h, k, l: Miller indices

Results and Discussion

The temperature dependence of the electrical resistivity (ρ) for $HgBa_2Ca_2Cu_3O_{8+\delta}$ at the pressure (0.2,0.3,0.5,0.6,0.9,1.0 and 1.1)GPa are shown in Fig. (1) It was found the behavior of the sample at P=0.2GPa. is semiconductor in the rang (300-80)K, while the other samples behave like a semiconducting phase in the rang (150-300)K or (200-300)K and then the behavior convert to a superconductor at temperature less than the range (150-200)K. In our opinion the sample prepared at P=0.2GPa has a transition temperature less than the boiling point of liquid nitrogen, our apparatus could not help as to obtain it.

It was found that the resistivity decreases with the increasing the pressure and the superconducting transition were not sharp and had tails around (120-80)K. This may be due to the fluctuation on the oxygen content or the existence of small amount of the secondary phase 1212 or 1201.

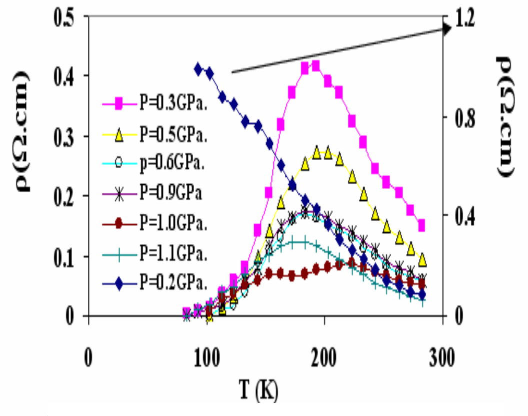

Fig. (1): Temperature dependence of resistivity for $HgBa_2Ca_2Cu_3O_{8+\delta}$ sintered at 860°C for various pressure in air

It is found from Fig. (2) and Table (1) that there is a variation of the transition temperature T_c with increasing pressure. In the range of P (0.3-0.5) GPa. there is an increase in the critical temperature, and T_c has a maximum value T_c=143K at P=0.5 GPa.. After that there is a decreasing in the critical temperature to 120 K at P=1.0GPa. and 1.1GPa.. The variation of T_c with increasing of pressure attributed to that, pressure induced change in carrier concentration assuming that the charge distribution among the crystallographically inequivalent CuO_2 layers is nonhomogeneous which are agreement with results of Mario Rabinowitz [11].

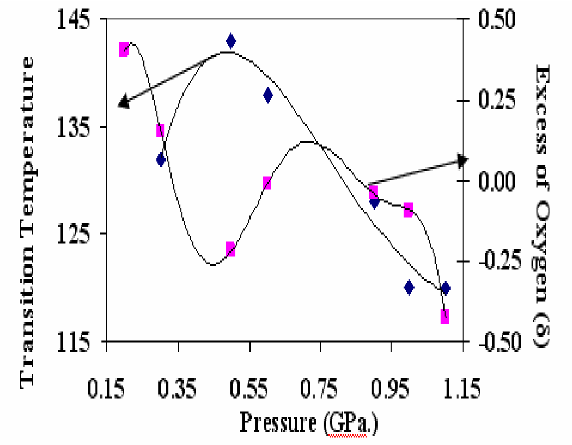

Fig. (2): Excess oxygen content (δ, transition Temperature (T_c) as a function of pressure

Table (1): Values of (δ) and T_c at different pressure

P(GPa.)	Excess of oxygen(δ)	T_c(K)
0.2	0.400	-
0.3	0.152	132
0.5	-0.217	143
0.6	-0.012	138
0.9	-0.043	128
1.0	-0.094	120
1.1	-0.420	120

Idometric titration was used to determine the oxygen content (δ) of the samples; the values of δ were listed in Table (1). It is interesting to note from this Table and Fig. (2) that there is a decreases of δ from 0.4 at P=0.2GPa.

to -0.4203 at P=1.1GPa., this will lead to a decreasing of holes in the perovskite layers and tend to decreasing of transition temperature.

The crystal structures of the prepared samples were studied by x-ray diffraction (XRD) analysis. X-ray diffraction patterns for all the specimens under study and Miller indices exhibited a tetragonal phase as shown in Fig. (3), with the lattice constants evaluated from 2θ of major peaks at different pressure are also listed in Table (2). The studied samples here exhibited two phases 1223, 1212 and the spectra of all samples include some impurity phases such as $CaHgO_2$ and CuO which are agreement with Akao *et al.* [12], this fact emphasis a polymorphic phase in the structure.

More than two phases is due to the displacement of an ion or oxygen defect or to the ordering of cations which lead to the stacking faults along the c-axis then this lead to deform the structure.

Table (2): Values of the lattice constant, volume cell and density at different pressure for $HgBa_2Ca_2Cu_3O_{8+\delta}$ system

P(GPa.)	a(A˙)	c(A˙)	c/a	V(A˙)^3	Density(gm/cm^3)
0.2	-	-	-	-	-
0.3	3.8759	15.9331	4.111	239.3581	6.063
0.5	3.8487	15.9724	4.150	236.5942	6.134
0.6	3.8445	15.9113	4.138	235.1722	6.171
0.9	3.8712	15.8767	4.101	237.9314	6.099
1.0	3.8508	15.8495	4.116	235.0278	6.174
1.1	3.8394	15.7731	4.108	232.5091	6.241

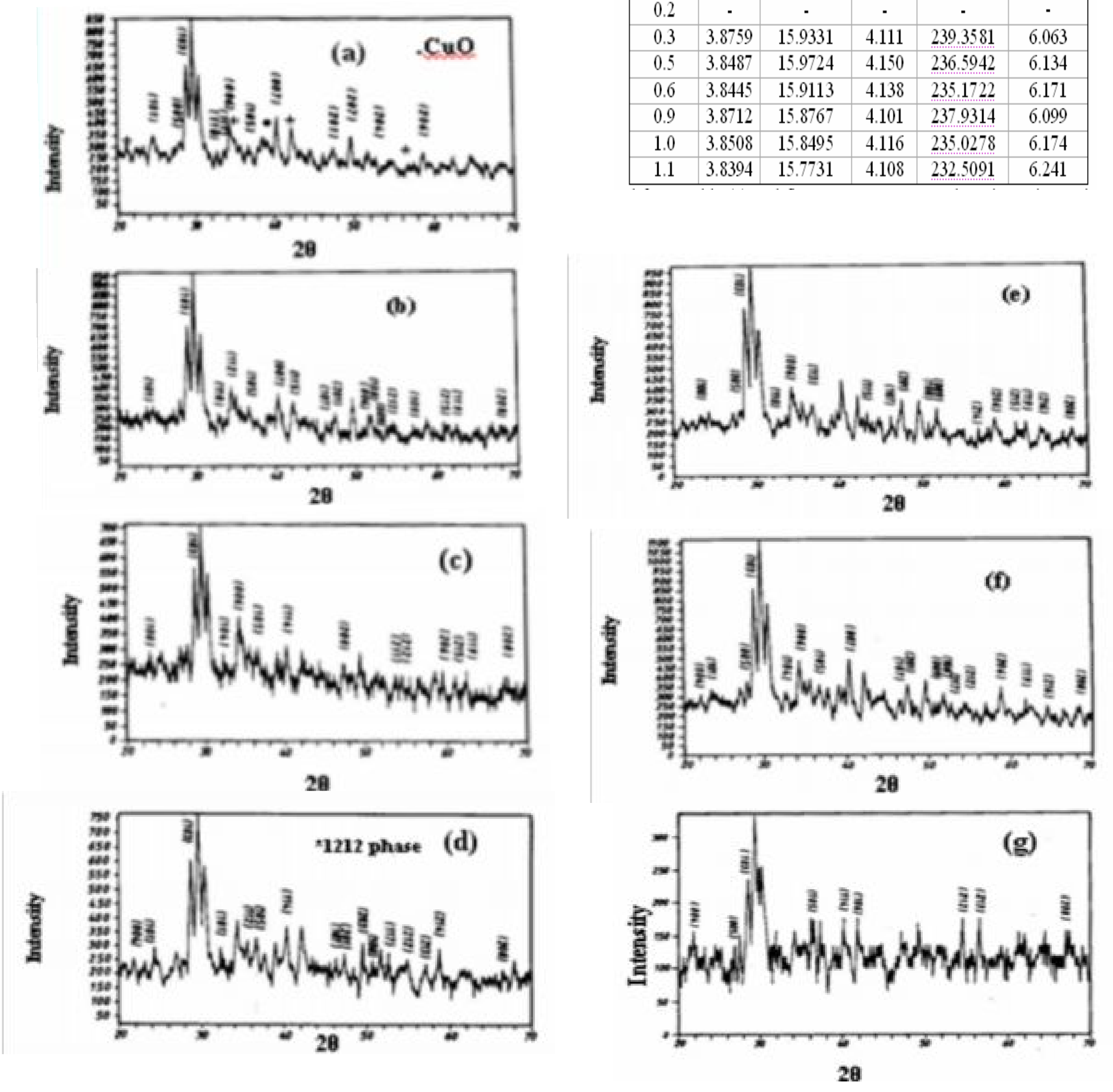

Fig. (3): X-ray diffraction patterns for the system $HgBa_2Cu_3O_{8+\delta}$ with different values of pressure, (a) P=0.2GPa., (b) P=0.3GPa., (c) P=0.5GPa., (d) P=0.6GPa., (e) P=0.9GPa., (f) P=1.0GPa., (g) P=1.1GPa.

Also we found from Table (2) and Figs. (4) and (5) that there is a decreases of the lattice constant (c) with the increasing of pressure from 0.5GPa.to 1.1GPa. On the other side, the lattice constant (a) and volume of the cell decreases with the pressure enhancement, except at pressure 0.9GPa. there is an increasing of them at this point. This may be due to deficiency of some atoms (oxygen defect, displacement of ions), adjusts the amount of charge transfer from Hg layer to Cu layer.

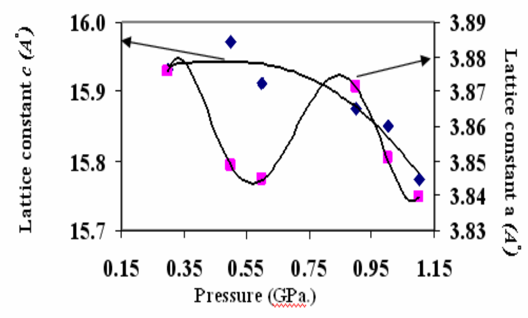

Fig. (4): Lattice constants as a function pressure

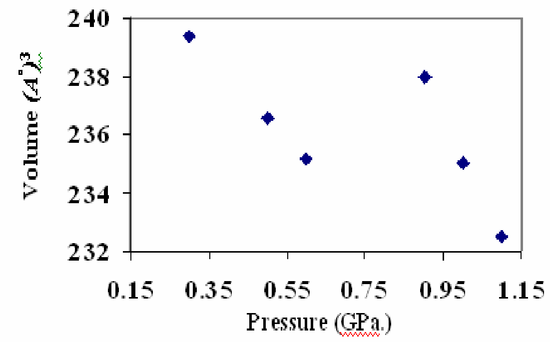

Fig. (5): Variation of volume cell as a function pressure

Beside that it was found that the value of density for the superconducting sample at different value of pressure, which are listed in Table (2). The value of density can be calculate from the equation [13]:

$$d = \frac{W_m}{N_A V}$$

Where N_A is the Avogadro's numbers in unit (particles/gm.mol), V is the volume of unit cell and W_m is the molecular weight in unit (amu).

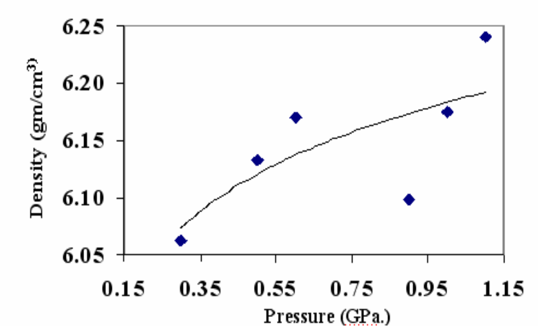

Fig. (6): Variation of density as a function pressure

From Table (2) and Fig. (6) we found that density increases from 6.063gm/cm^3 at P=0.3GPa. to 6.241gm/cm^3 at P= 1.1GPa.

Conclusions

1. It has been observed from the resistivity measurements:
   A- The behavior of the system $HgBa_2Ca_2Cu_3O_{8+\delta}$ at P=0.2 GPa.is semiconductor in the rang (300-80)K while the behavior for other values of pressure convert to a superconductor at temperature less than 150K.
   B- T_c increased with the increasing of pressure from 0.3-0.5 GPa. the maximum T_c=143K is at P=0.5 GPa. then T_c decreases to 120K at P=1.1 GPa.
2. X-ray analyses have showed a tetragonal phase and there is a decreasing in c-axis lattice constant with the increasing of pressure.
3. Densities are increases with the increasing of pressure.

References:

[1] Rajiv Giri, H.K. Singh, R.S. Tiwari and O.N. Srivastava ”Bull. Mater. Sci,” V24, No.5, 2001, pp.523-528.

[2] S.N.Putilin, E.V. Aptipov, O. Chmaissem, M. Marezio ”Nature” V362, 1993,p.226.

[3] Q.L. Xu, T.B. Tang and Z.J. Chen “Supercond. Sci. Technol.” V7, p.828, 1994.

[4] H.M. Chao, L.J. Chen, J.C. Chen, X.Y. Hua, P.F. Yuan and X.X. Yao "Physica C”, V232, 5,1994.

[5] L. Jansen and R. Block, ”Physica A” V212, p.143, 1994.

[6] N.H. Hur, N.H. Kim, K.W. Lee, K.H. Yoo, Y.K. Park and J.C. Park "PhysicaC" V234, p.19, 1994.

[7] X. S. Wu, H. M. Shao, S. S. Jiang, C. Gou, D. F. Chen, D. W. Wang, Z.H. Wu. ” Physica C V: 261, Issue: 3-4, May 1,pp189-195. 1996.

[8] E.J. Gonzalez, W. Wong-Ng, G.J. Piermarini, C. Wolters and J. Schwartz ”Powder Diffraction” V12, pp106-112, 1997.

[9] E. V. Antipov, J. J. Capponi, C. Chaillout, O. Chmaissem, S. M. Loureiro, M. Marezio, S. N. Putilin, A. Santoro and J. L. Tholence "Physica C"V218, pp348-355, 1993.

[10] H.Bougrine,M.Houssa,R.Cloots,M. Pekalal,I.Sargankova and M.Ausloos ” *Supercond. Sci Technol* “. V11 pp.128-132, 1998

[11] M.Rabinowitz and T.McMullen “Irainbow@stanford.edu.

[12] T.Akao,S.R.Lee,K.Mizogami,H.Suematsu and H.Yamauchi “Physica C" V338,pp.76-83,2000.

[13] L.H.Van Vlack, “Elements of materials science and engineering”, 6th edition. Addition-Wesley Publishing Company, Inc. (1989).