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Theory and Experiments: SCF Calculations/ XRD/Enhanced Scattering/PLD in YBCO7-d Nano Film

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III. Theory and Experiments: SCF Calculations/XRD/Enhanced Scattering/PLD in $YBCO_{7-d}$ Nano Film

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ABSTRACT:

X-Ray diffraction above and below the transition temperature to superconductivity T_c are correlated using SCF-MOLECOLE computations that describe the CuO_2 layer (fig. 1) in the field of 9 unit cells of $YBa_2Cu_3O_7$. The MO symmetry, charge transfer and O_AO_B bond mobility are compared to the data using the highest occupied molecular orbitals, MO_{-i} (HOMO=MO_0) bases to construct the layer tight-binding extended state.

1. INTRODUCTION

SCF calculations[1] of the CuO_2 conduction plane in cuprates are used to ascertain the consistency of and Cu-$L_{2,3}$ edge energy[2] shifts observed experimentally below T_c in films and bulk crystals in hope to identify phenomena associated with superconductivity.

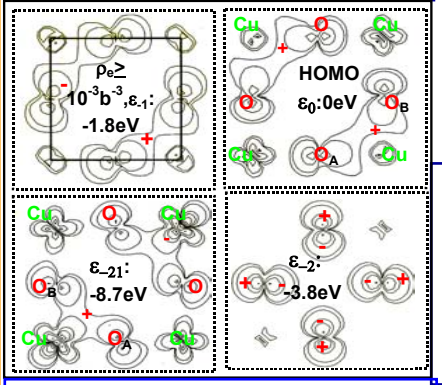

Figure 1: Maps of electron density for some of the highest occupied Cu_4O_4 MO with $\rho_e \geq 10^{-3}b^{-3}$, sign of the amplitude and diagonal O_A-O_B sigma bonding. Excitation of electrons occupying the HOMO, changes the electron density at two diagonal Cu sites from $\pm 3d_{x2-y2}$ symmetry and $s_z=\pm 1/2$ weakly bonded, to $\pm 3p_{xy}$ in MO_{-1}, MO_{-2}. At lower energy MO_{-21} shows not a square but a prevalent CuO_2: O_A-Cu-O_B triangular bonding electron density[1].

2. Discussion of SCF calculation/experiments/ on nano-film $(CuO_2)_n$ layer covalent properties to describe PLD source/effects:

The MOLECOLE program developed by Gina Corongiu and Enrico Clementi[1a] in a LINUX operating system was used to obtain the SCF energy levels and electron density of different size $(Cu_4O_4)_xM_y$ lamella in the charge field of 9Q unit cells (Table I), where M are charge compensating ions in adjacent layers above and below the CuO_2 planes, as a function of the total charge and periodic lattice distortions, PLD observed in YBCO films[1b,c].

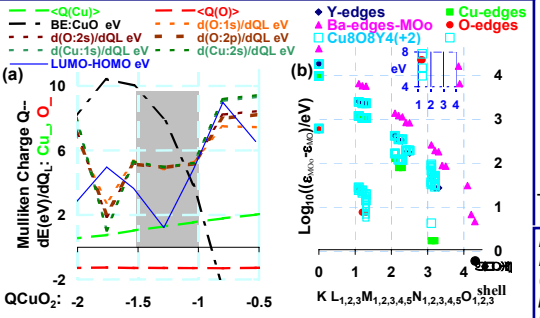

Figure 2: MOLECOLE SCF results for lamella in the point charge field of 9 YBCO unit cells using the bases: Cu/O: GaussianGEOSMALL, Y:Gaussian11. (a) $(Cu_4O_4)_2$:148±4 MO SCF calculations determine the Mulliken charge Q, Cu-O bond BE, O and Cu core edge energy variations indicating stability in shaded area versus $-1.5<QCuO_2<1$ where Q point charges≈Mulliken Q(Cu), Q(O) obtained in repeated iterations. (b) $(Cu_4O_4)_2Y_4$: 225 MO compared to atomic edge energies. Insert shows that the Ba $O_{2,3}$-shells can and do interact with MO_{21} (fig. 1) as indicated by the non-zero overlap in z-direction and Ba $M_{4,5}$ edge shifts to lower E below T_c[1c] indicating a valence decrease.

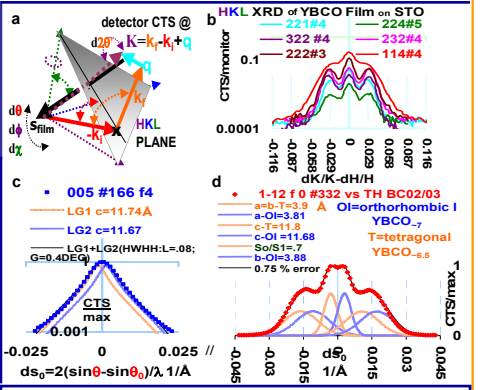

Figure 3. XRD of YBCO film on $SrTiO_3$[4] at E=8.048keV: (a) H,K,L≠0, dH=-dK,dL=0 XRD with sidebands $sb_{\pm 1}$ that obtain q_{ab} = $(dK/K\text{-}dH/H)_{sb\pm 1} \approx 2^{-1/2}/24$. (b) XRD measurement parameters showing how the transfer of momentum $q \| k_F$ may lead to the observed PLD. (c, d) 005 and 1-12 XRD fitted to two equal phase fractions in equilibrium:

$$2(YBCO_{7,\,metal})_{n2} \Leftrightarrow X^{\neq} \Leftrightarrow (YBCO_{6.5,\,insulator})_{2\,n2} + n^2\, O_{interstitial}$$

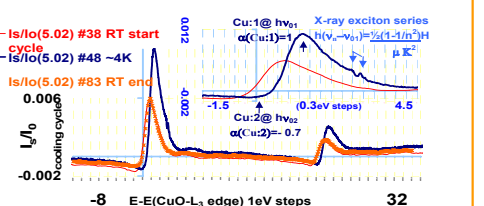

Figure 4: I_s/I_0 [2] extremes of reversible cooling cycle versus E-E(CuO,Cu:L_3-edge=931.2eV), E_{Bragg}-E=150-135eV[4c,7c]. The baselines coincide, but at 4 to10K the edge energy shifts by 0.65±0.05eV with respect to room temperature. Insert's expanded scale shows structure vs T, and how enhanced scattering I_s/I_0 by atoms at Cu:i sites are identified by the opposite sign of α(Cu:i, i=1,2). Excitonic series is observed.

3. Conclusions:

SCF calculations and measurements made at SSRL and ALS synchrotron laboratories describe the properties of fabricated $YBCO_x$ soft solid nano-films. A path for the metal insulator transitions (fig. 3) between phases that are also found in bulk YBCO. The intermediate $X^{\neq}$ may be associated with $(O_A{:}O_B)_{1/2q}^{-2}$ bi-radical chains formed when changing the direction of diagonal conductivity by X-ray flux effects and/ or by temperature T near T_c[3].

1.(a)J.V. Acrivos and O. Stradella, *International Journal of Quantum Chemistry*, **46**, 55(1993); **(b)**M. A. Navacerrada and J.V. Acrivos, NanoTech 2003, **1**, 751 (203); **(c)** H.S. Sahibudeen, M. A. Navacerrada and J.V. Acrivos, NanoTech 2005, **2**, 573 (05) **2.**J.B. Kortright et al., *J. Magn. Magn. Materials*, **207**, 7 (1999). **3.(a)** N.F. Mott, *"Metal Insulator Transitions"*, Taylor and Francis, ltd, 1974, 1990; **(b)** A.S. Alexandrov and N.F. Mott, *"Polarons and Bipolarons"*, World Scientificific, Singapore (1995); **(c)** G. Koster, T.H. Geballe, and B. Moyzhes, *Phys. Rev.* **B66**, 085109 (2004).