LETTER TO THE EDITOR

Cellular automata model for persistent photoconductivity in YBCO

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Abstract. We feel that persistent photoconductivity has lacked an adequate microscopic model to date. In an effort to remedy this, we present a cellular automata model based on the idea that trapped electrons in the basal plane lead to structural rearrangements when illuminated with light ranging from IR–UV wavelengths. We interpret our results within a single-band model for the electronic structure of YBCO and find that our results are in good agreement with previous experimental work.

1. Introduction

Persistent photoconductivity (PPC) and persistent photo-induced superconductivity (PISC) are two remarkable phenomena exhibited by the family of superconductors $YBa_2Cu_3O_{6+x}$ (YBCO) [1] as well as Tl-based compounds [2]. In YBCO, the superconducting properties (i.e. diamagnetism, T_c) can be tuned over a wide range by illumination with light. Understanding the mechanism for PPC and PISC may help unlock the mechanism for high-temperature superconductivity [3].

Recent experimental results have suggested that persistent photoconductivity in $YBa_2Cu_3O_{6+x}$ may be explained by a model which incorporates both electrons trapped at oxygen vacancies, and resulting structural rearrangement in the basal plane [4]. Persistent photoconductivity is characterized by a long-lived photodoped state which shows no sign of relaxation below 250 K [5]. Accompanying the large relative changes in resistance ($\Delta\rho/\rho \sim 100\%$ or more) are transport and structural changes. During illumination, the c-axis is observed to contract, with the same functional dependence on time as the resistivity [6]. Additionally, the bond length distribution between the Cu(1) and apical O(4) sites is observed to split into two well defined peaks during illumination [7]. During illumination, heavily oxygen deficient material near the insulator–metal transition may even become superconducting [5]. A study of the enhancement of T_c by illumination [8] shows that, as a function of oxygen content, the net change in T_c is similar to the room temperature annealing effect studied by Veal *et al* [9]. Because of these parallels, PPC was viewed by some as simply due to basal plane oxygen rearrangement. The causes for such rearrangement were not specified, but were believed to be related to the intrinsic properties of the material. A few concrete models have been proposed, mainly in the form of percolation calculations.

One model combines this approach with the asymmetric next-nearest neighbour Hamiltonian to calculate the enhancement of T_c as a function of oxygen content. In this model,

incident photons are thought to create perturbative dipole moments leading to lengthening of the chains [10]. Another approach treats the chains as metallic islands embedded in an insulating matrix [11]. A percolation threshold is calculated for low oxygen content (6.15) and the crossover to overall metallic behaviour in the chains is observed (6.35) near the insulator–metal transition. The infrared (IR) optical properties are compared before and after illumination. The changes that are seen in the IR spectra are opposite to those seen when the samples are heated without changing the oxygen content. The chain lengths can be inferred from other methods, and the optical properties after illumination are consistent with the average chain length in the basal plane increasing.

Other experimental results cannot be explained by structural rearrangements in the basal plane. Changes in the photoluminescence spectra are seen to be consistent with the population of defects after illumination [12]. Also, the infrared photoconductivity changes character after illumination with visible light [4, 13]. That is, after illumination with visible light, and a reduction in resistance, illumination with IR wavelengths causes the resistance to increase. This phenomenon is known as IR quenching. The model proposed to explain these results suggests that electron–hole pairs are created during illumination and that electrons are trapped at oxygen vacancies while holes are transferred to the CuO_2 planes to contribute to the conductivity. However, IR quenching only succeeds in recovering about 5–10% of the resistance change induced by visible light [13], hampering a purely trapping model's ability to account for the large change in resistance upon illumination.

Remarkably, PPC has been observed in over-doped compounds such as $Tl_2Ba_2CuO_{6+x}$ (Tl2201) [2]. In contrast to YBCO, the changes induced by visible illumination are completely reversible by illumination with infrared light. In this material there is no analogous structure to the chain layers in YBCO, so the mechanism for PPC is thought to be defect based [2]. A thorough review of PPC in high-T_c materials including experimental results and proposed mechanisms may be found in [3].

Given the experimental results discussed above for YBCO as well as Tl2201 we have come to believe that any successful model for PPC will include both defects as well as structural rearrangements. In furtherance of this aim, we have developed a simple cellular automata model in order to represent the effect of local perturbations on the average chain length, and in turn the resistivity. In our model, these local perturbations are trapped electrons at oxygen vacancies. The application of some simple rules based on local occupancy and other factors leads to growth of the chains. We will attempt to interpret our results within a one-band model [14]. To our knowledge, this is the first such attempt at a microscopic model for PPC in which the population of defects results in the observed structural rearrangements, and so represents an important step forward in understanding the fundamental PPC mechanism.

2. Model

For our current purposes, we are interested in the narrow range of oxygen contents around the insulator–metal transition. This is where the absolute change in resistivity is largest when oxygen deficient YBCO is illuminated [14]. Our thought experiment involves illumination of $YBa_2Cu_3O_{6.3}$ at low temperature (< 100 K) until saturation of the PPC effect. Presumably, the sample is cooled from room temperature at a moderate rate (0.1 K s^{-1}) and we do not expect any oxygen order/disorder effects analogous to that observed in the O–II phase [9]. The resistivity is monitored at or near DC frequencies during illumination. We represent the basal plane by a square array shown in figure 1. For reasons of computational efficiency, we have assigned the Cu(1) sites to even–even coordinates in terms of the ordered pair (Y,Z) shown on figure 1. Oxygen vacancies are assigned to even–odd, and odd–even coordinates.

Odd–odd sites are not occupied, as these are interstitial sites. The oxygen content is fixed at the start and vacancies are filled randomly. Since the simulation is designed with low-temperature illumination experiments in mind, the oxygen is considered to be 'frozen' in place. Once the oxygen sublattice is populated, the mean chain length is computed. The distribution of various chain lengths in our model are consistent with previously reported calculations [14]. We typically chose an array size that included $N \times (N + 1)$ oxygen sites, a fraction, x, of which are occupied. Here, x is the oxygen content in $YBa_2Cu_3O_{6+x}$.
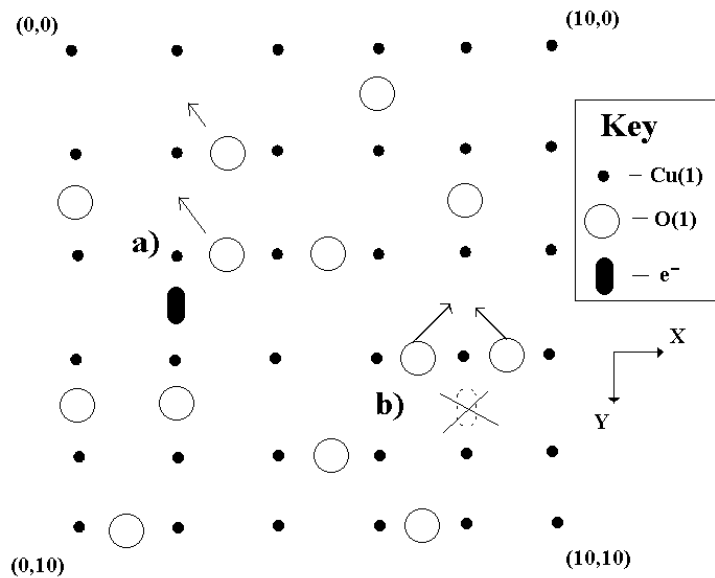

Figure 1. Representation of basal plane in tetragonal YBCO: (a) an electron trapped at the site indicated causes the motion of oxygen into chain, and disturbance is able to propagate to next lattice site and (b) an electron cannot be trapped at a site because both oxygen atoms would like to move to a vacancy in the chain.

Once the array has been built up, oxygen vacancies are chosen at random and perturbed. We associate each perturbation with a unit of time. We feel that this is justified because the magnitude of the PPC effect has been shown to depend only on the cumulative photon dose [5]. At fixed flux, each absorbed photon produces an electron–hole pair, so the number of perturbations and time are directly proportional. Once a vacancy has been chosen, the local occupancy is scanned and an 'electron' can be trapped provided the following conditions are met: (1) the trapping of an electron results in the motion of oxygen into the chain layer, (2) there is a vacancy for the oxygen to move into, and (3) there is no more than one trapped electron per unit cell. Figure 1 gives two illustrative examples for consideration. Once the initial trap event has occurred, the disturbance is allowed to propagate along the chain (Y), proceeding until a vacancy is encountered (figure 1(a)). Alternatively, both diagonal sites could be filled ($X \pm 1$) in which case the disturbance would not be allowed to propagate (figure 1(b)), and therefore the electron would not be trapped. In this fashion, we see the trapping event as requiring a balance of long- and short-range forces. First, the electron exerts a local influence on the Cu(1) sites. This disturbance is passed along, propagating only along the chain directions, causing an instability which allows for reordering of the oxygen atoms. After each trapping event, the average chain length is computed, with the electron considered to bridge adjacent Cu(1) sites.

3. Results and analysis

The average chain length is recorded as a function of the number of perturbations. Figure 2 shows the results for several lattices with 50×51 unit cells. The average chain length rises steeply at first, and then levels off quickly, characteristic of stretched exponential growth. We have fit the chain length to the following form:

$$l(pert) = l_\infty - \Delta l \cdot \exp\left[-\left(\frac{pert}{pert_{ill}}\right)^\beta\right]. \tag{1}$$

The change induced by 'illumination' is $\Delta l = l_\infty - l_0$, $pert$ is the number of perturbations, and $pert_{ill}$ is a constant obtained from the fit (analogous to τ_{ill} in [5]). The average chain length before the lattice is perturbed is l_0 and the final value is l_∞. For the several lattices shown in figure 2, $\Delta l/l \approx 65$–70%. Typically, β varies between 0.75 and 0.85. The fit to equation (1) with $\beta = 0.84$ for one of the lattices is shown in the inset of figure 2. For the reader unfamiliar with PPC, we point out that during illumination with light, the resistivity decreases according to stretched exponential decay in a functional form similar to equation (1) [1].
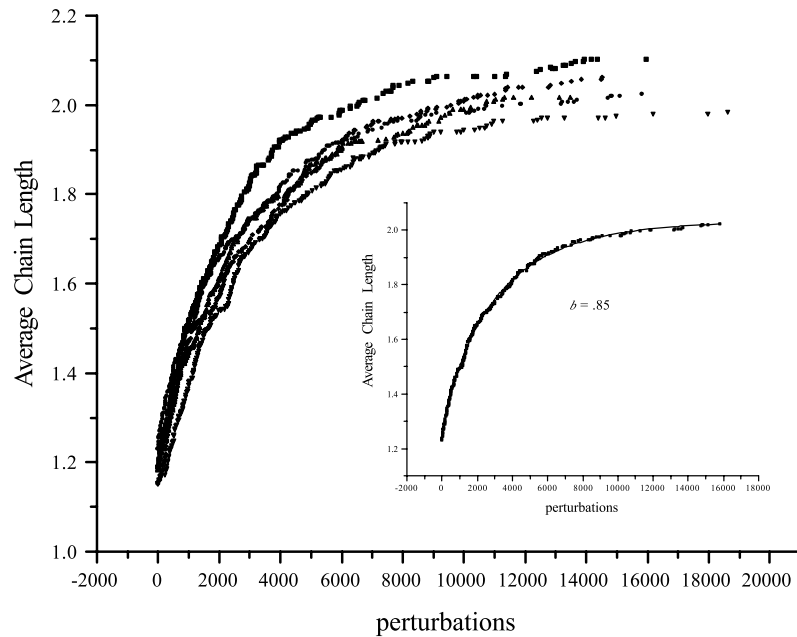

Figure 2. Growth of average chain length with the number of perturbations. Inset shows fit of equation (1) to one of the lattices.

3.1. Single-band model

In a one-band model for the electronic structure of YBCO, relative changes in resistivity are given by:

$$\frac{\Delta \rho}{\rho} = -\left(\frac{\Delta \mu}{\mu} + \frac{\Delta p}{p}\right). \tag{2}$$

The hole density is p, and we will define it as the number of holes per formula unit CuO_2. As a starting point for analysis, we think it reasonable to assume $\rho \sim 1/l$ where l is the average

chain length, as will be justified below. The changes in resistivity due to illumination will be accounted for in the following way. First we will count the number of chains of greater than the minimum length necessary to 'inject' holes into the CuO_2 planes (i.e. $l \geqslant 3$). It has been theorized [15] that chains greater than this critical length transfer holes to the puckered planes. Furthermore the amount of holes transferred is proportional to the length of the chains with the proportionality factor being around 0.7 [15]. However, the possibility is held out that the proportionality factor may be closer to 0.4 or 0.5 than 0.7 for shorter chains. Finally, in order to calculate $\Delta p / p$, we must know the hole density before illumination. For $YBa_2Cu_3O_{6.3}$ we expect p to be 0.026 in accordance with [16–18].

Next we turn our attention to the increase in average chain length, Δl. We would like to directly correlate this with an increase in mobility. We think that such an association is reasonable because optical measurements on crystals show that increasing the chain length (oxygen content) results in a lower scattering rate, and hence mean free time [19, 20]. Extrapolating the infrared conductivity measurements of [20] to near DC frequencies shows that the carrier effective mass is not dramatically different for fully oxygenated YBCO as it is for oxygen deficient material. Conversely, the scattering rate for YBCO with several different oxygen contents is very similar in the mid-infrared, but is very different near DC frequencies [20]. The net result is that the mobility is enhanced by increasing the chain length. Within our range of current understanding the only significant changes that occur as the result of adding oxygen are an increase in the average chain length and an increase in the hole density in the planes. Therefore, in accounting for our results, we feel that the association $\Delta l / l \simeq \Delta \mu / \mu$ is appropriate.

We would like to comment on the proportionality factor governing the amount of holes transferred from the chains to the planes. According to [15] this occurs for chain fragments greater than length 3. We note that the factor controlling the transfer of holes out the chains may be closer to 0.5 for short chains ($n \leqslant 6$) [15]. It is stated quite clearly that the use of the factor 0.7 is valid in the limit of infinitely long chains. In the case that we use 0.5, we see about a 10% reduction of $\Delta p / p$ than if we use 0.7. The vast majority of chains that develop are less than length 6, so this is not a terribly critical point.

For the five lattices that we have considered the average relative change in the mobility, $\Delta \mu / \mu = 0.7$. The average increase in hole density, $\Delta p / p = 0.35$. So, in accordance with a single-band model, $\Delta \rho / \rho \simeq -1.1$. This is in good agreement with our previous experimental results [4] as well as those of others [14]. Our results are in rough agreement with a study in which the increase in mobility dominates the increase in hole concentration for a $YBa_2Cu_3O_{6.35}$ thin film [2]. In the work of Tanabe *et al* the change in mobility outweighed the change in hole concentration by a factor of about 2.5 [21]. This ratio is smaller for us, approximately 2.1. Additionally, their results show that the relative increase in hole density is independent of oxygen content. We predict a larger relative change in the hole density than they do. This discrepancy may be related to the fact that we are just below the metal–insulator transition, while they were right on the metallic side. We note that our predicted increase in CuO_2 plane hole concentration is right on the threshold for superconductivity, and overall metallic behaviour. Since the hole concentration drops off rapidly below the insulator–metal transition, a small increase should lead to a much larger relative change. This would help explain the extremely large $\Delta \sigma / \sigma$ (2.9) for a $GdBa_2Cu_3O_{6.1}$ sample reported in [14]. When $\Delta \sigma / \sigma$ is plotted against oxygen concentration for $RBa_2Cu_3O_{6+x}$ ($R = Y$, Gd), the increase in $\Delta \sigma / \sigma$ below the metal–insulator transition is said to be 'faster than exponential' [14]. Based on our model, we would predict that this is due to the large relative change in hole concentration, and that further below the metal–insulator transition ($x < 0.3$) the increase in hole concentration will dominate the increase in mobility.
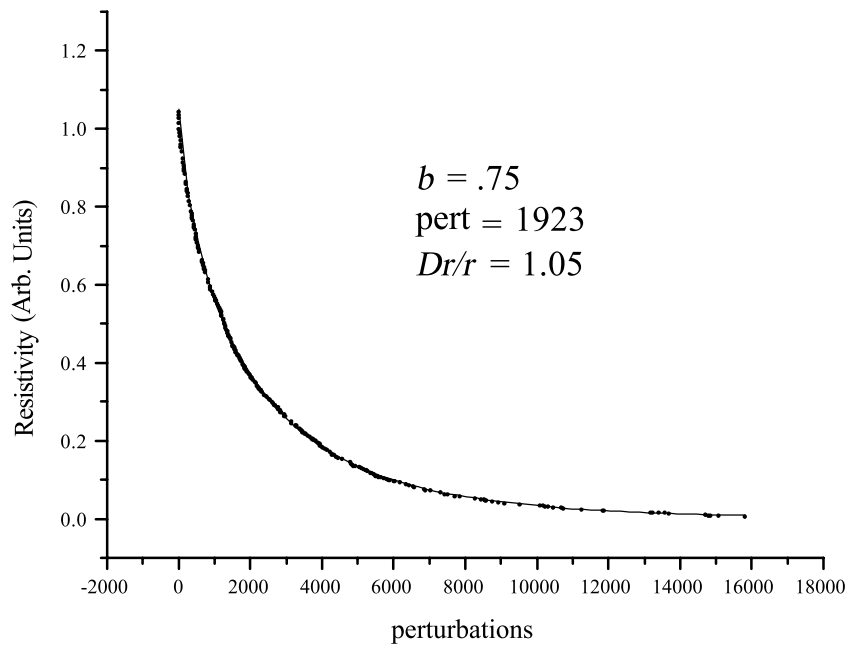
Figure 3. Simulation of resistivity as a function of illumination time resulting from assumption that $\rho \sim 1/l$ and $\Delta\rho/\rho = \Delta\mu/\mu + \Delta p/p$. Points are generated by our model, solid line is a fit to equation (1) with chain length replaced by resistivity, as is experimentally observed.

As a final step, we have developed a functional for the resistivity as a function of illumination time, or perturbations. This is shown in figure 3, where the fit parameters appear on the graph. The dependence on perturbations is clearly that of a stretched exponential with $\beta = 0.75$. Our results are applicable in the limit of a weak flux, in that only one disturbance propagates at a time. It will be interesting to see how the exponent varies with the number of simultaneously propagating perturbations. We are developing a more thorough model in which there will be several disturbances propagating at once. Additionally, we would like to extend our results to all oxygen contents. We are also interested in applying our model to the resonant raman scattering (RRS) results recently observed [22].

4. Conclusion

We have presented a simple cellular automata model for PPC in oxygen deficient YBCO. Using a single-band model, we are able to account for the magnitude of the effect as well as the stretched exponential dependence of the resistivity on illumination time. We present these results in support of our earlier experimental work [4]. We favour a model for PPC in which the population of defects leads to structural rearrangements in the basal plane. We propose applying our model to the entire range of oxygen contents as well as new observed phenomena.

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