The Role of Defects in Persistent Photoconductivity in YBa$_2$Cu$_3$O$_{6+x}$

J. F. Federici$^{1}$ and Daniel M. Bubb$^{1,2}$

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In the persistent photoconductivity (PPC) phenomena, illumination of a material with light leads to a long-lived photoconductive state. During illumination of oxygen deficient YBCO near the insulator-metal transition, the illuminated material may even become superconducting. Accompanying the large relative changes in resistance are transport and structural changes as well as changes in the photoluminescence and infrared quenching spectrum. Infrared quenching data show that the magnitude of IR quenching saturates quickly as a function of visible photon dose, long before the PPC effect saturates. Our experimental and theoretical results have suggested that persistent photoconductivity (PPC) in YBCO may be explained by a model that incorporates both electrons trapped at defects (oxygen vacancies) and resulting structural rearrangement in the basal plane. A simple cellular automata model based on the idea that trapped electrons in the basal plane lead to structural rearrangements when illuminated with light predicts the correct time dependence of the photoexcitation process.

**KEY WORDS:** photoconductivity; superconductivity; quenching; defects.

1. INTRODUCTION

Several years ago the phenomena of Persistent Photoconductivity (PPC) [1] and Photoinduced Superconductivity (PISC) [2] were first reported in oxygen deficient YBCO. Persistent photoconductivity is characterized by a long-lived photodoped state. Accompanying the large relative changes in resistance ($\Delta \rho/\rho \sim 100\%$ or more) in the normal state are transport and structural changes. During illumination, the $c$-axis is observed to contract, with the same functional dependence on time as the resistivity [3]. 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 [4]. During illumination, oxygen deficient material near the insulator-metal transition may even become superconducting. Resistance, mobility, optical, and structural experimental studies and theoretical explanations of the PPC and PISC effects have recently been reviewed [5].

Attempts to explain the mechanism have generally fallen into two classes: (1) photoassisted oxygen ordering [6], and (2) a defect mechanism [7–10]. In the defect mechanism, an absorbed photon of light generates an electron–hole pair. The photogenerated electron is trapped at a defect—presumably an oxygen vacancy [11,12]. The trapping of an electron prevents the recombination of the hole with the electron, resulting in the transfer of a hole to the CuO$_2$ planes where it contributes to the conductivity. PPC has also been observed in overdoped compounds such as Tl$_2$Ba$_2$CuO$_{6+x}$ (Tl2201) [13]. 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 [18], with the role of oxygen vacancies being replaced by that of interstitial oxygen.

In the photoassisted oxygen-ordering model, photons enable basal plane oxygen atoms to move...
within the lattice and become part of the CuO$_2$ chains. As these chains lengthen, they inject holes into the CuO$_2$ planes by acting as charge reservoirs [14]. A study of the enhancement of $T_c$ by illumination [15] 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. [16]. Likewise, the time dependence of the resistivity changes and corresponding energy barriers are similar for PPC and oxygen ordering. Because of these parallels, PPC was viewed by some as simply due to basal plane oxygen rearrangement [5].

Given our optical, transport, and structural experimental results for YBCO, we have come to believe that any successful model for PPC will include both defects as well as structural rearrangements. In particular, the trapping of photogenerated electrons at defect sites is a precursor to structural rearrangement.

2. EXPERIMENTAL DETAILS

The YBa$_2$Cu$_3$O$_{6+x}$ samples used in this study were either fabricated at the Army Research Laboratory (ARL), the Naval Research Laboratory (NRL), or Advanced Technology Materials (ATM). Samples were grown either by pulsed laser deposition (PLD) or by single liquid source metalorganic chemical vapor deposition (MOCVD) with the details of the film growth, annealing, and characterization published elsewhere [8–10]. Typical samples studied had an oxygen content in the range of $x = 6.2$–$6.4$. Samples are attached to a cold finger and placed in an optical cryostat. The samples are cooled and fully immersed in liquid nitrogen (LN$_2$). For each run, before illumination, the sample’s resistivity versus temperature curve is recorded. A current is applied across a bias resistor and a differential voltage across the sample is measured using standard 4-point resistivity techniques.

3. EXPERIMENTAL EVIDENCE FOR DEFECTS

3.1. Luminescence

If defects are important for PPC and PISC, then perhaps the defects luminesce and the luminescent intensity changes during exposure to light as the defects (traps) become filled. Experimental results [8] showing a correlation between photoluminescent intensity and the PPC state as well as a correlation between the photoluminescence spectrum and the wavelength dependence of the photoinduced conductivity suggest that defects play a role in PPC in which the defects act as weakly luminescent F-centers under illumination. Theoretical treatments of defects in YBa$_2$Cu$_3$O$_{6+x}$ have shown that stable F centers can form in the CuO$_2$ chains [17]. Moreover, it has been shown that changes in the c-axis and local orthorhombic symmetry can be directly attributed to electron trapping in the chain layers or the conversion of an O$^{2-}$ ion into O$^-$ [18].

As an example, Fig. 1 shows the time dependence of the resistivity and 1.88-eV luminescence during the 4-W illumination of a YBa$_2$Cu$_3$O$_{6.4}$ sample. The correlation between the photoinduced conductivity and

![Fig. 1.](image-url)
Role of Defects in Persistent Photoconductivity 333

Luminescence strongly suggests that the luminescence at 1.88 eV is indicative of the PPC/PISC state. The change in luminescence at 1.88 eV under 4-W illumination is observed to be consistent with its assignment as a measure of the number of available oxygen vacancies. While an insulating YBa$_2$Cu$_3$O$_{6.4}$ sample shows the largest change in resistance (∼37%) and 1.88-eV luminescence (∼15%) upon 4-W illumination, a YBa$_2$Cu$_3$O$_{6.8}$ sample shows smaller decreases in resistance (∼19%) and 1.88-eV luminescence (∼6%). No perceptible change is observed in the resistance or the luminescence spectra of YBa$_2$Cu$_3$O$_{6.5}$ superconducting samples, since there are no oxygen vacancies available to trap photogenerated electrons. These observations are consistent with recent luminescence measurements which show that luminescence can be used as an indicator of charge transfer from the chain layer to plane layer in YBa$_2$Cu$_3$O$_{6.5}$ [19].

In comparing the PPC efficiency spectrum in the visible range [20,21] with known photoluminescence peaks, there appears to be a strong correlation between the two spectra, which suggests that both the luminescence spectrum and the PPC efficiency spectrum have a common origin. However, the strong PPC efficiency peak near 4.1 eV does have a corresponding photoluminescence spectrum and the PPC efficiency spectrum has several practical applications such as optically controlled superconducting switches, microbridges, in situ laser fabrication/tuning of SC circuits [9,22,23], storage and recording applications. One difficulty in using the PISC mechanism for optically controlled fast switching of YBa$_2$Cu$_3$O$_{6.5}$ thin films is that the photoinduced state is long-lived. However, by using a combination of visible and IR light sources, one may contemplate optimally controlled superconducting circuit components that can be tuned on a time scale of tens of seconds.

3.2.1. Experimental Results

We have observed IR quenching of the photoinduced state for wavelengths with energies less than 1.6 eV. For the typical data shown in Fig. 2, the sample (immersed in LN$_2$) is illuminated with visible light until the PPC effect is nearly saturated. After the visible light is turned off, illumination of the photoinduced sample with low intensity infrared light increases the resistance, as shown in Fig. 2. The maximum change in resistance ∆R shown in Fig. 2 for 980 nm corresponds to about 2.4% of the resistance drop induced by the 4-W illumination. A value of ∆R% = 100 corresponds to complete quenching of the photoinduced state. The quenching process is reversible: After infrared quenching, illumination with argon light causes the sample resistance to decrease [9]. This process can be repeated, suggesting the possibility of optically controlled superconducting devices if the IR quenching efficiency could approach 100%. The IR quenching spectrum [8] is shown in Fig. 3 for several samples. The ordinate, quenching efficiency, is defined as the ratio of the change in resistivity due to illumination with IR light to that of the change in resistivity due to illumination with visible light. The largest quenching (∼5–10%) was observed for 0.38-W broadbanded infrared light (1.5–5 μm) [9]. This suggests that a broad distribution of defects is responsible for PPC. (The longer wavelength range was limited by the spectral transmission of the cryostat’s windows.) Experiments are currently underway to extend IR quenching band to beyond 10-μm light.
Fig. 2. (a) Time dependent resistance during argon ion laser illumination of YBCO sample immersed in LN2. At $t = 60$ s the sample is exposed to visible laser light. At $t = 3600$ s light is blocked; (b) Time dependent change in resistance of sample upon illumination with 980-nm (top curve) and 148-nm light (bottom curve) once visible light is blocked.

Fig. 3. Comparison of photoinduced absorption spectrum ([24, 25], solid lines) to IR quenching spectrum (dashed lines). The upward arrows correspond to individual data points [15]. The dashed line connecting those points is included as a guide to the eye. The downward arrow indicated data (dashed line) above 1.26 eV [16]. The similar onset wavelength and width of spectrums suggest a common origin.

Figure 4 shows the intensity dependence of IR quenching (the change in resistivity $\Delta R$) subsequent to illumination with infrared light at 1300 nm (0.954 eV). Replotting the data from Fig. 4 versus total photon dose shows that the total flux of IR photons determines the change in resistance. The dependence on cumulative dose is analogous to what is observed for excitation of PPC/PISC. Furthermore, the dose dependence suggests a one-electron process.

Another interesting effect is the response of the oxygen deficient samples before and after exposure to visible light. As shown in Fig. 5, the resistivity decreases during exposure to 887-nm light before exposure to visible light. In the PISC efficiency measurements [20] reported by Kudinov et al., the response at 1.4 eV was negligible compared to that at 2.4 eV. Our results contradict this. After exposure to visible light in order to induce PPC, illuminating the sample with 887-nm light causes an increase in resistance.
Role of Defects in Persistent Photoconductivity

These results are consistent with our earlier measurements [9], which showed a finite PPC effect for photon energies in the IR. Apparently, the PPC efficiency spectra extends not only through the ultraviolet [5], but also down through the infrared to at least about 2 μm.

The PPC response to IR wavelength saturates at a lower photon dose than the one at visible wavelengths. We suggest that this occurs because there are a smaller percentage of the total defects being populated, that is, only those with energy equal to and less than 0.93 eV are populated when illuminating with 0.93-eV light. When illuminating with photon energy above 1.6 eV, the total available defects are populated and the response to IR light changes.

We undertook a study [8] of the magnitude of quenching as a function of visible photon dose (Fig. 6). The behavior of the samples is quite surprising in that the character of the response to IR changes dramatically for photon doses as low as ~10¹⁸ [18]. The response to IR light is not appreciably different when it has been exposed to >10 [25] photons as opposed to 10 [18] photons. This change suggests that defects populate quickly, compared to the other dynamics of the PPC effect. Contributing to this interpretation is the photomodulation spectroscopy/photoinduced absorption [24,25] work done by Wei et al. For these measurements the fractional changes in the IR transmission spectra between 0.09 and 2.5 eV is measured during low intensity visible illumination. They observed two distinct energy bands, one broad (~1 eV) and centered at 0.8 eV called the high energy (HE) band. They observed another band having width 0.1 eV at 0.1 eV, called the low energy (LE) band. Additionally the absorption bleaches above 1.6 eV and saturates around 1.3 eV. We observe the onset of IR quenching at 1.6 eV and saturation around 1.3 eV. A comparison of the photoinduced absorption spectra and IR quenching spectra shown in Fig. 3 shows that both measurements give the same onset energy near 1.6 eV and a broad spectral dependence, suggesting that the two effects have a common origin and therefore are manifestations of the same phenomenon.

3.2.2. Interpretation

In interpreting the IR quenching results, it seems that there are two roles that defects may play in PPC for YBCO. First, they may be an independent phenomenon associated with chain oxygen vacancies in YBCO. As a distinct mechanism of PPC, their contribution may be at most 10% compared to oxygen ordering [15] as inferred from the fact that IR quenching
Fig. 6. $\Delta R$ per IR photon as a function of visible photon dose. The ordinate changes quickly from large and negative to positive values. The saturation of $\Delta R$ for low visible photon doses suggests that the filling of the defects occurs quickly during the photoexcitation process for PPC.

can only cause a small change in resistivity when compared with that caused by visible light. However, it seems unlikely to us that the mechanism responsible for IR quenching and PPC are unrelated. In particular, they have overlapping spectral features: The PPC efficiency spectra overlaps with the IR quenching spectra such that wavelengths above about 1.6 eV preferably reduce the resistance, while wavelengths below 1.6 eV generally increase the resistivity. While that alone may not be convincing, the fact that the overlapping spectral features depend intimately on one another is suggestive. IR wavelengths initiate PPC before exposure to visible light and later partially quench it, suggesting to us that the mechanisms for both phenomena are intimately related.

A more comprehensive description of PPC should include defects as precursors to oxygen ordering during illumination with visible light. The trapped charges modify the local electric field distribution, which leads to induced dipole moments. These dipoles create the necessary impetus for the movement of basal plane oxygen [26,27] with consequent lengthening of chain fragments. Once the chain fragments become greater than the critical length, they inject holes into the CuO$_2$ planes and the bulk conductivity is enhanced.

Since the PISC/PPC effect persists for hours at 77 K, the trapped electron (F-center) must be an extremely stable configuration. Two features stabilize the F-center and prolong the lifetime of the photogenerated carriers: lattice deformation around the trapped electron and physical separation of the electrons and holes. Similar to other materials with F-centers, one expects that the lattice locally deforms around the trapped electrons [17]. Lederman et al. [28] report that upon photoexcitation of YBCO, the lattice parameters shrink, indicating a more stable configuration. Changes in the lattice parameters can be attributed to electron trapping [18]. In addition, the large spatial separation between photogenerated electrons, which are trapped at oxygen vacancies in the CuO chain, and holes, which are in the conductive CuO$_2$ planes, implies a small overlap of their wavefunctions and therefore a lower probability for recombination of electrons and holes to quench the photoinduced conductivity.

In our model PPC would result from the combined action of two mechanisms. First, upon photoexcitation, defects are quickly populated with trapped electrons resulting in changes in the local electric field structure. Secondly, as a result of the modification of local fields, structural changes would occur over a
much longer time scale, which reflect the periodicity of the lattice. Then, PPC is understood as the sum effect of an electronic contribution (defects—fast) and a lattice contribution (ordering—slow). This model can account for the quicker saturation of IR quenching as well as the much smaller change in resistance when compared to PPC. Further, this view on the role of defects is consistent with almost any model that contains oxygen ordering and actually enhances them by providing a clue as to the precursor of this ordering.

When oxygen deficient YBCO is illuminated with IR radiation, the response to IR wavelength is dramatically different, depending on whether or not the sample has been exposed to visible light. Before the sample is exposed to visible light, IR light initiates the PPC state. After the sample is exposed to visible light, IR light partly quenches the PPC state. Oxygen ordering alone cannot account for the existence of IR quenching, nor the fact that it saturates for one part in 10^5 of the photon dose required for saturation of the PPC effect. The IR quenching efficiency is not substantially different after illumination with 10^18 photons as opposed to 10^23. Were this simply a manifestation of the PPC effect, one would expect the IR quenching efficiency to be proportional in some way to the visible photon dose. However, the magnitude of IR quenching does not appear to be related to the cumulative visible photon dose unless the functional form is on a scale vastly smaller than the PPC effect. In that case, this result would still be suggestive of two phenomena on different time scales. Different time dynamics exhibited by IR quenching and PPC in YBCO, coupled with the observed wavelength dependence of IR quenching, suggest that defects may play an important role in PPC.

Once the array has been built up, oxygen 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 [30]. We typically chose an array size that included N = (N + 1) oxygen sites, a fraction, x, of which are occupied. Here, x is the oxygen content in YBa2Cu3O6+x. Details of the model are given elsewhere [7].

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 [30]. Our thought experiment involves illumination of YBa2Cu3O6+x at low temperature (<100 K) until saturation of the PPC effect. The resistivity is monitored at or near DC frequencies during illumination. 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 [30]. We typically chose an array size that included N × (N + 1) oxygen sites, a fraction, x, of which are occupied. Here, x is the oxygen content in YBa2Cu3O6+x. Details of the model are given elsewhere [7].

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4. MODELING OF PHOTOEXCITATION PROCESS

The thermal relaxation process of PPC and its stretched exponential time dependence is well understood [5,29] as resulting from a distribution of energy barriers or relaxation times. In fact, it has been shown by Phillips [29] that the β parameter of a stretched exponential decay is not an adjustable parameter but rather can only assume certain values. Until recently, the physical origin of the time dependence of the photoexcitation process was unclear [5]. Recently, we developed a simple cellular automata model [7] for PPC in which the trapping of photogenerated electrons at defects results in structural rearrangement of the oxygen. The model predicts the effect of trapped photogenerated electrons (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 and a corresponding decrease in resistivity with illumination, which follows a stretched exponential law. The magnitude of induced resistivity changes and other parameters are in good agreement with experimental data.

4.1. Cellular Automata Model

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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. Once the initial trap event has occurred, the disturbance is allowed to propagate along the chain, proceeding until a vacancy is encountered. Alternatively, both diagonal sites could be filled in which case the disturbance would not be allowed to propagate, 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.

4.2. Results and Analysis

The average chain length is recorded as a function of the number of perturbations. Figure 7 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:

\[ l(\text{pert}) = l_\infty - \Delta l \cdot \exp \left( \frac{\text{pert}}{\text{pert}^{\text{ill}}} \right)^{\beta} \]  

The average chain length before the lattice is perturbed is \( l_\infty \) and the final value is \( l_1 \). The change induced by “illumination” is \( \Delta l \). pert is the number of perturbations, and pert\(^{\text{ill}}\) is a constant obtained from the fit. For the several lattices shown in Fig. 7, \( \Delta l/l = 70–72\% \). Typically, \( \beta \) varies between 0.8 and 0.9. The fit to Eq. (1) with \( \beta = 0.84 \) for one of the lattices is shown in the inset of Fig. 7.

In analyzing the results for a predicted change in resistivity versus perturbations, the changes in the chain length can be related to the change in mobility \( \beta \) by \( \Delta l/l = \Delta \mu/\mu \). The mobility can be related to the resistivity via a one-band model:

\[ \frac{\Delta \rho}{\rho} = - \left( \frac{\Delta \mu}{\mu} + \frac{\Delta p}{p} \right) \]  

For the five lattices that we have considered, the average relative change in the mobility is \( \Delta \mu/\mu = .72 \). The
average increase in hole density, $\Delta p/p = .35$. So, in accordance with a single-band model, $\Delta p/p = -1.1$. This is in good agreement with our previous experimental results [8] as well as those of others [30].

Using the one-band model, the resistivity as a function of illumination time, or perturbations, can be calculated as shown in Fig. 8. The dependence on perturbations is clearly that of a stretched exponential. However, we do note that it is necessary to compare our results with films whose entire volume is excited by light. Our results are applicable in the limit of a weak flux, in that only one disturbance propagates at a time.

As part of our assumptions for the model, after each trapping event, the average chain length is computed, with the electron considered to bridge adjacent Cu(1) sites such that trapped electrons are considered to be part of the chain length. This may give some indication as to why there appears to be two different time scales and magnitudes for IR quenching compared to visible initiation of the PPC effect. Upon visible illumination, photogenerated electrons are quickly trapped. On a longer time scale, there is oxygen rearrangement in the basal plane that lengthens the chain fragments. Upon IR illumination, a portion of the trapped electrons are freed which then recombine with the holes. The time scale of this process (electronic) should be comparatively fast when compared to structural rearrangement of the basal plane. Consequently, there should be a fast partial recovery of the resistivity with IR illumination. Moreover, when trapped electrons are freed by IR illumination, the rearranged chain oxygens do not immediately revert back to their initial (unilluminated) positions because of a fairly large energy barrier ($\geq 1$ eV) for its motion at low temperature. Consequently, there is only a partial recovery of the visibly initiated persistent photoconductivity with IR illumination.

5. CONCLUSION

The role of defects in the persistent photoconductivity (PPC) phenomena for YBCO is described. Based on optical, transport, and structural measurements a model of PPC is suggested, which incorporates both photogenerated electrons trapped at defects (oxygen vacancies) and resulting structural rearrangement in the basal plane in which the trapping of electrons is a precursor to structural rearrangement. The model correctly accounts for the experimental behavior of IR quenching. A simple cellular automata model based on the idea that trapped
electrons in the basal plane lead to structural rearrangements when illuminated with light predicts the correct time dependence of the photoexcitation process as well as the correct size of the changes in resistivity with illumination.

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