Plastic electronic devices fabricated from carbon based molecules and polymers\cite{1} have received considerable academic and industrial attention over the last decade.\cite{2} They offer the promise of flexible and low cost broad area devices, which can be fabricated using high speed, low temperature roll-to-roll production techniques.\cite{3} Devices such as organic light emitting diodes (OLED) displays/lighting\cite{4,5}, organic photovoltaic (OPV) devices,\cite{6} organic field effect transistors (OEFTs),\cite{7} and photodetectors\cite{8–11} have been demonstrated. However, organic semiconductor materials are inherently more disordered than their inorganic counterparts,\cite{12} due to a lack of a regular crystalline lattice. Thus charge carrier recombination rates are generally higher, while mobilities are generally around three to four orders of magnitude lower.\cite{13} This means that, although organic devices offer the potential for lower cost devices, performance is in general not as good as their inorganic counterparts.

Recently there has been considerable attention focused on understanding the relationship between mobility,\cite{13} recombination,\cite{14} and the distribution of carrier transport states within organic semiconductors\cite{15} with the objective of improving carrier lifetime/transport and thus device performance. Methods such as transient photovoltage,\cite{16} photo charge extraction by linearly increasing voltage (photo-CELIV),\cite{17} and impedance spectroscopy\cite{18} have been applied to obtain mobility and recombination data. Charge carrier lifetimes in the range of 1 ps–0.3 s have been reported from devices fabricated from both solution processed polymers\cite{19–23} and evaporated small molecules.\cite{24–28} These measurements have in general been performed on working devices where the band edge of the metallic contacts (typically aluminum and Indium tin oxide (ITO)) has been aligned to the highest occupied molecular orbital (HOMO)/lowest unoccupied molecular orbital (LUMO) of the semiconductor to increase carrier injection and extraction efficiency. However, the equilibrium Fermi level of the organic semiconductor is usually mid way between the HOMO–LUMO level, and far from that of the ITO or aluminum contact. This means for the system to reach equilibrium, and for the quasi-Fermi levels of the metal and semiconductor to align, a significant number of both electrons and holes must be injected from the contacts into the bulk of the organic semiconductor, resulting in a large equilibrium background carrier population. The nongeminate recombination rate ($R$) in organic semiconductors is known to be a strong function of both local electron ($n(x)$) and local hole carrier ($p(x)$) densities,\cite{29,30} $R(x) = k(x,n,p)n(x)p(x).$\cite{29,30}$ $ where $k$ is a constant of proportionality and $x$ is the position. Thus if the local background charge density of either species could be reduced, so would the recombination rate. Furthermore, if photogenerated electrons could be quickly swept from hole rich regions, and photogenerated holes from electron rich regions, the spatial overlap of the photogenerated populations would be reduced and charge carrier lifetime would also be increased.

In the following pages we demonstrate that by engineering the band structure in a plastic electronic device, hole carrier lifetimes longer than 1 hour can be achieved. We attribute this four order of magnitude increase in carrier lifetime to a hill shaped band structure which forces photogenerated electrons and holes to spatially separate shortly after generation, the reduction in spatial overlap between the electron and hole population reduces the recombination rate. The hill shaped band structure also generates a hole rich and electron poor region in the center of the device in which the hole recombination rate is low, this acts as a charge storage area. These results demonstrate that it is possible for organic semiconductors to have long charge carrier lifetimes, and opens up the possibility for new classes of ultra low light level photodetectors and memory elements.

Figure 1a depicts the device structure; it consists of an ITO coated glass substrate with contact resistance 30 Ω sq$^{-1}$. An insulating layer of SiO$_2$ is spin coated from a sol-gel onto the ITO film, followed by a layer of the organic semiconductor N,N’-Di[(1-naphthyl)-N,N’-diphenyl]-1,1’-biphenyl]-4,4’-diamine (NPB). Finally a 30 nm thick aluminum capping electrode is evaporated onto the NPB. Full details of device fabrication can be found in the Supporting Information. Charge is stored within the device by photocexcitation through the transparent ITO contact using a 355 nm laser with a 10 ns pulse.
length and a fluorescence of 110 µJ cm\(^{-2}\). The flat band structure of the device is depicted in Figure 1b and the chemical structure of NPB is shown in Figure 1c. To measure the charge stored within the device, a positive voltage ramp of duration \(t_{\text{extract}}\) and magnitude \(V_{\text{max}}\) is applied to the ITO contact. This voltage ramp is applied at a time \(t_{\text{delay}}\) after the optical excitation ends. The timings of the voltage transients are shown in Figure 2a.

Two typical current transients measured from the system are shown in Figure 2b (black and red lines) along with the corresponding voltage ramp applied to the device (blue line). The black line represents the current measured from the device when no laser excitation is applied, while the red line shows the current transient when the device is photoexcited 1 ms before the start of the voltage ramp. In both cases it can be seen that when the applied voltage starts increasing at time \(t=0\), the current produced by the device rapidly increases reaching a plateau of 0.014 mA within 10 µs. This plateau of current corresponds to the geometric capacitive current being drawn out of the device from the contacts. Note, charge density was calculated by dividing the measured charge from the device by the volume of the NPB active layer. When no photoexcitation is applied to the device (black line), the current remains stable until 400 µs when the voltage ramp ends. For the case where the device is photoexcited (red line), a Gaussian shaped bump of charge appears on the transient starting at 50 µs and ending by 200 µs. It has a maximum height of 0.03 mA at 90 µs. The area under this Gaussian of charge corresponds to the charge photogenerated within the device. It should also be noted that a positive bias is applied to the ITO contact, meaning that the extracted current is due to holes.

Figure 3a plots current transients as a function of delay time between photoexcitation and extraction \(t_{\text{delay}}\). The black line represents charge extracted from the device after 100 µs, while the blue line represents charge extracted from the device after 1 h. Figure 3b plots the integrated photogenerated charge from Figure 3a as a function of time, it can be seen that after 1 hour around 25% of the photogenerated charge still remains in the device, suggesting that electron–hole recombination rate is much slower than previously observed in organic electronic devices.

Figure 3b-inset plots the photogenerated charge extracted from a device when the SiO\(_2\) carrier blocking layer is replaced with a polystyrene (PS) blocking layer. It can be seen that this device has a much higher recombination rate than the device with the SiO\(_2\) blocking layer, indeed the charge density decreases by an order of magnitude over the first 100 ms. To investigate if the ultra long charge carrier lifetimes in the SiO\(_2\) sample are due to an interface effect or a bulk effect, SiO\(_2\)/NPB devices were fabricated with a thin PS layer separating SiO\(_2\) from the NPB. When the PS layer was thin (19 nm) long charge carrier lifetimes were still observed, however when the thickness of the PS layer was increased charge carrier time decreased—see Figure 3c. 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potential within the bulk of the device is key to determining long charge carrier lifetimes. To further support this assertion, we fabricated devices with an insulating layer consisting purely of poly(methyl methacrylate) (PMMA). PMMA has a relatively high permittivity of \( \varepsilon = 4.9 \), and so if the distribution of electrostatic potential within the bulk of the device is important, one would also expect to also see long lived charge carrier lifetimes in this device. This was in fact the case, and we were able to extract charge tens of minutes after photoexcitation—see the Supporting Information for details.

Previously carrier lifetimes in organic systems have been observed in the range 1 ps–0.3 s, thus observing carriers with a lifetime more than four orders of magnitude more than previously reported was not expected. If the current transient obtained after photoexcitation in Figure 2b is examined (red line) it can be seen that until around 50 ps after the voltage ramp has been applied no photogenerated charge is extracted. Only once the applied voltage has reached the value of 1.68 V (at 50 µs) can the photogenerated charge be accessed. The need for an applied bias of over one volt to extract photogenerated charges suggests that there is an energetic barrier between where the charge is stored in the device and the extracting contact.

The presence of an energetic barrier hindering charge extraction suggests the presence of a Schottky contact and associated band bending separating the stored photogenerated charge within the device from the extracting contact. Schottky contacts and band bending in electronic devices are associated to regions of space charge, which in turn is associated with material doping. Although the NPB layer is not intentionally doped with water, oxygen, or nonpure materials can lead to unintentional and relatively high levels of background doping in organic devices.

From Figure 3c we can see that by changing the material used in the blocking layer from SiO\(_2\) to polystyrene the charge carrier lifetime can be significantly reduced. The choice and relatively high levels of background doping in organic devices have been reported to cause unintentional doping effects, which can lead to unintentional and relatively high levels of background doping in organic devices.

Finally, it should also be noted that the current transients measured from this device upon the application of a voltage ramp are very different in nature to those observed from organic solar cells, which are fabricated not to have an energetic barrier between the contacts and where charge is generated. Thus in organic solar cells there is no delay between the application of the voltage transient and extraction of photogenerated charge.

To better understand why the device can store charge for long periods of time with very little recombination occurring, and why an applied bias of over 1 V is required to extract the stored charge, we model the device using a time domain drift-diffusion model. To calculate the electrostatic potential distribution between the ITO contact (\( x = 0 \)) and the Al top contact (\( x = d \)), Poisson’s equation

\[
\frac{d}{dx} \varepsilon_x \varepsilon_r \frac{d\phi}{dx} = q (n_f + n_i - p_f - p_i - N_{ad})
\]

is solved over the SiO\(_2\) layer and the NPB semiconductor. Where, \( \phi \) is the position dependent potential, \( \varepsilon_x \) is the permittivity of free space, \( q \) is the charge of an electron, \( N_{ad} \) is the density of dopant molecules, and \( n_f / p_f, n_i / p_i \) are the densities of free and trapped electrons/holes, respectively. To account for drift and diffusion of charge carriers within the device, the bipolar drift diffusion equations are solved over the same region

\[
J_+ = q \mu_e n_f \frac{\partial E_{LUMO}}{\partial x} + qD_n \frac{\partial n_f}{\partial x}
\]

\[
J_- = q \mu_h p_f \frac{\partial E_{HOMO}}{\partial x} - qD_p \frac{\partial p_f}{\partial x}
\]

where \( J_+ / J_- \) are the electron and hole current densities, \( E_{LUMO}, E_{HOMO} \) are the energy levels of the LUMO and HOMO, respectively, and \( \mu_e, \mu_h \) are the electron and hole mobility.

Figure 3. a) Charge extraction transients with varying times between photoexcitation and charge extraction (\( t_{\text{delay}} \)). The current features after 400 ms is discussed in the Supporting Information. b) Black squares: integrated photocharge extracted from the device as a function of \( t_{\text{delay}} \) corresponding to Figure 3a, note that it takes over an hour for the charge density to decay to 25\% of its initial value. Red stars: Integrated charge from the device when the SiO\(_2\) blocking layer is replaced with a polystyrene blocking layer. By changing the blocking layer the carrier lifetime is significantly reduced. c) Charge carrier lifetimes for a ITO/SiO\(_2\)/PS/NPB/Al device where the thickness of the PS layer is varied.
moieties. Nongerminate recombination and carrier trapping are described using the Shockley–Read–Hall recombination model.\textsuperscript{[32]} Exponential carrier trap states between the HOMO and LUMO mobility edges are assumed.\textsuperscript{[13]} The transfer matrix method is used to calculate the photon distribution within the device. A full description of the device model can be found elsewhere.\textsuperscript{[34,35]} It should also be noted that we assume charge is generated within NPB in accordance to the process described in ref. \textsuperscript{[36]}. The SiO\textsubscript{2}/NPB sandwich structure is modeled, by using two layers of semiconducting material. The insulating SiO\textsubscript{2} layer is assumed to be transparent and given a low mobility of \(1 \times 10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}\) to prevent conduction of charge. While the NPB layer is assumed to have an optical absorption coefficient of \(1 \times 10^{-11} \text{ m}^{-1}\) as measured using UV–vis spectroscopy (see the Supporting Information) and a mobility of \(1 \times 10^{-6} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}\), a full list of simulation parameters can also be found in the Supporting Information. The interface between the sol-gel deposited SiO\textsubscript{2} and NPB is assumed to have a 2 eV step (discussed in the Supporting Information).

**Figure 4.** a) Band bending as a function of doping. As more doping is introduced the bands bend more. b) The corresponding current transients, it can be seen as doping increases the transients start to resemble more those of the experimental transients. The same line colors to denote doping have been used in both figures. The dopants are considered p-type because charge is extracted on application of a positive voltage.

![Figure 4](image-url)

**Figure 5.** Photogenerated charge remaining in the device as a function of time. It can be seen that when the device is undoped (red line) almost all the photogenerated charge recombines quickly. As doping is increased (green line), carrier lifetime increases. When the doping level is further increased (blue line), no recombination can be observed within 1000 µs after photoexcitation. The dots represent the case where the energetic barrier between the SiO\textsubscript{2} and NPB is removed allowing charges to penetrate the SiO\textsubscript{2}, as could happen due to the presence of defect states.

![Figure 5](image-url)
electrons and holes attempt to minimize their energy. Thus the electrons drift down towards the contacts and become extracted (indeed upon photoexcitation a negative current spike can be seen, representing the photogenerated electrons leaving the device—see the Supporting Information for details). While the holes drift towards the center of the device and become trapped by the potential profile of the device and the SiO₂ insulator layer. This process results in spatially separated electrons and holes. Because there is no spatial overlap between the electron and hole populations, charge carriers cannot recombine. Another important feature of having upwardly bent bands is that in the center of the device where the photogenerated charge becomes trapped, the Fermi level is close to the HOMO, producing a hole rich-electron poor region, which will have a low hole recombination rate.

Figure 7a plots the simulated charge extraction process as the positive voltage ramp is applied to the left hand side of the device. The top panel plots the HOMO band structure as a function of increasing applied bias. It can be seen that as the applied potential is increased the highest point of the HOMO is pulled down, eventually reducing to the same height as the right hand extracting contact. Underneath the plot of the HOMO, the hole density as a function of position is plotted. It can be seen as the positive potential is applied to the device the photogenerated charge packet moves from the left hand side to the right, always trying to minimize its energy. At later times (not shown) the packet is fully extracted from the device. Figure 7b summarizes the charge extraction process in pictorial form, the photogenerated charge can be seen being pushed from the left hand side of the device to the right.

Above, we suggested that the difference in lifetimes observed between the SiO₂ device and the PS device could be due to the different permittivities of the layers ($\varepsilon_{\text{SiO}_2} = 3.9$ and $\varepsilon_{\text{PS}} = 2.5$). This observation can be better understood by examining Figure 8a which plots simulated charge carrier lifetime as a function of carrier blocking layer permittivity. It can be seen from the figure that the higher the relative permittivity, the longer the lifetime of the charge carriers. This can be explained by examining Figure 8b where the band structure at equilibrium is plotted as a function of relative permittivity. The higher
the relative permittivity of the insulating layer, the smaller the potential drop over the insulating layer and the further away the LUMO is from the equilibrium quasi-Fermi level. This will mean that for high permittivities, there will be few electrons and thus a low recombination rate in the region of the device where photogenerated charges are stored.

Figure 9a plots the experimental current transients as a function of maximum applied voltage using a delay time of 10 µs. It can be seen that as the maximum voltage ($V_{\text{max}}$) is increased, the peak in the current due to the photoexcited charge shifts to the left and becomes narrower. The simulation of this process is depicted in Figure 9b. That the simulation can predict these experimental trends, suggests that the model is a good approximation to the physical processes occurring within the device.

Detecting ultra low power optical signals, such as light from distant stars, is important in science, medical, and defense applications. The ability of the structure to produce long-lived photogenerated charges opens up the possibility to use the device in a time-integrating mode, whereby a very low photon flux over long periods of time cause the gradual build up of charge. Once enough charge carriers have been collected to make the signal detectable by the external electronics, charge could be extracted. To demonstrate this, an ITO/SiO$_2$(105 nm)/NPB(70 nm)/Al structure was constantly illuminated at 365 nm using a low brightness source over exposure times ranging from 1 to 7000 s. At the end of the measurement the voltage ramp was applied to the device to extract the charge. The experiment was performed at light fluxes of 1.6, 3.2, 4.8, and 6.4 µW cm$^{-2}$. The results are plotted in Figure 10. It can be seen that, as expected, a gradual buildup of charge is observed over time.

Figure 8. a) Charge carrier lifetime as a function of carrier blocking layer permittivity. b) The corresponding band structure.

Figure 9. Experimental a) and simulated b) charge extraction transients as a function of $V_{\text{max}}$. If the experimental and simulated results are compared, it can be seen in both data sets, that as $V_{\text{max}}$ is increased the current peak shifts to the left and narrows.

Figure 10. Charge build up in the device on exposure to a constant low light flux. In this mode the device is operating as an ultra low light level photodetector.
Another potential use of ultra long lived charge carriers could be in optically writable electronically readable memory elements. Charge could be stored in the device with a light pulse and the device’s state measured or reset by applying the voltage ramp. In Figure 3b we demonstrated that charge can be stored in this device for more than 1 h, however it should be noted that we expect longer lifetime to be accessible with a higher quality SiO₂ layer, controlled doping of the semiconductor layer, and possibly by using an insulating layer with a higher permittivity than SiO₂.

We have demonstrated an organic electronic device in which we observe carriers with a lifetime of over 1 hour. Carriers with lifetimes at room temperature in disordered organic semiconductors exceeding 0.3 s have not been previously been reported. Using a combination of experimentation and simulation, we attribute this long lifetime to band bending which drives electrons and holes apart, spatially separating them within the device shortly after photogeneration. The holes are then stored in a hole-rich electron-poor region of the device. We demonstrate that by carefully tuning the energetic landscape of the device carrier lifetimes can be significantly increased. This principle opens up the possibility of developing a new generation of plastic electronic ultra low level photodetectors and memory elements.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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