Light-induced transient currents from molecular films in a tunneling microscope junction

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(Received 20 February 1998; revised manuscript received 20 August 1998)

Under pulsed light, transient currents are observed between a thin molecular film and the metallic electrode on which it is deposited, when a tunneling microscope tip acts as a counterelectrode. We interpret this effect in terms of photo-assisted charge injection to or from trap states in the molecular film. The bias and time dependence of the transients provide information on the energy distribution and dynamics of the charge. [S0163-1829(99)07403-2]

I. INTRODUCTION

The charging of insulators is a venerable subject, but many aspects of it are still not well understood. This is not so surprising, because we cannot apply many techniques of theory and experiment that serve us well in dealing with transport in metals and semiconductors. However, it is clear that the kinds of techniques we most need are those that investigate charge transport at or very near the surface of an insulator or the interface between an insulator and a conductor. Charge will transfer if a metal and an insulator are brought into contact. This is the phenomenon of ''contact electrification'' (for a review, see Ref. 1). Similarly, if an insulating film is deposited on a conductor, charge will generally transfer across the interface. Electrons from the conductor can tunnel to states near the interface and, depending on the degree to which the states are localized, migrate deeper into the insulating film.

In this work we describe studies in which visible-wavelength light causes charge to move into or out of a molecular film. This effect is quite different from photopopulation/depopulation, which has been extensively studied in insulators (primarily silicon dioxide, because of its great technological importance). See, for instance, Refs. 2–4. Let us briefly review photopopulation/depopulation, considering an insulator in terms of band structure with a forbidden gap greater than about 7 eV. Ultraviolet light is used to inject charge, promoting electrons from the Fermi level of a metallic conductor, E_F, to conducting states of an adjacent insulator (e.g., Ref. 5). Electrons may therefore move quite far in the insulator and subsequently fall into trap states lying somewhere below the conduction band. This is the process of photopopulation. Later, the traps are emptied, again via the conduction band, by a variable-wavelength light source and an applied bias sweeps the freed charge to the electrode. Note that in these experiments, without light or thermal excitation there is no communication between the electrode and the traps, so filled traps above E_F can remain filled indefinitely. Similarly, empty traps below E_F can remain empty indefinitely.

In the present experiments we believe that the metal film electrode and the traps in the molecular films communicate primarily via tunneling processes. Electrons in these deep traps cannot be activated to conducting states of the insulator with the available light energy and certainly not with thermal energy. However, because the traps are very close to the interface, electrons can transfer directly to or from the conductor by photoassisted or potential (''conventional,'' non-photoassisted) tunneling processes. We shall not explicitly discuss transport between traps or include it in the model we shall use, as the observed transient behavior can mostly be explained without considering it. It is nonetheless clear that such transport must exist and there is great interest in the processes involved. For a discussion of these issues and spectroscopic measurements of transport between traps using a direct electron injection technique, see Ref. 6. In our work we use visible light, the energy of which is sufficient only to assist the tunneling of electrons into and out of trap states near the metal film electrode with energies lying less than approximately 2.5 eV above E_F. Thus, in contrast to photopopulation/depopulation, the equilibrium situation in our experiments in the absence of light is that traps above E_F will be empty and traps below E_F will be filled. For a discussion of the kind of tunneling process that we believe maintains this equilibrium situation see Ref. 7. The application of light causes some population of traps above E_F and some depopulation of those below E_F. Our measurements provide information about the populations of the traps and the relaxation of those populations.

The samples of interest here are usually, but not always, organic molecular films deposited on a gold film. The sample is mounted in a scanning tunneling microscope (STM), the tip of which is slightly flattened and acts as a counterelectrode to the gold film. In many transport situations in which the conducting path is constricted, fluctuating occupation of traps is responsible for ''random telegraph noise.'' This noise is often seen in STM currents and therefore in STM images.[8] In order to isolate and analyze the behavior of a very few traps, STM studies of the statistics of current noise have been performed for the Si/SiO_2 interface[9] and for molecular films.[10] However, to study dynamics one would like to actively probe the traps. Although it is possible to use our technique to map spatial variations in the trapped charge density by using a scanning mode, we will concentrate here on information obtainable at a fixed position using fairly-long-time signal averaging. By measuring transient currents resulting from repetitive trap depopulation and repopulation we can obtain time-dependent information limited only by the bandwidth of our electronics. As we shall mention later, in future experiments using optical multiple-pulse techniques, even this limitation should be surmountable.

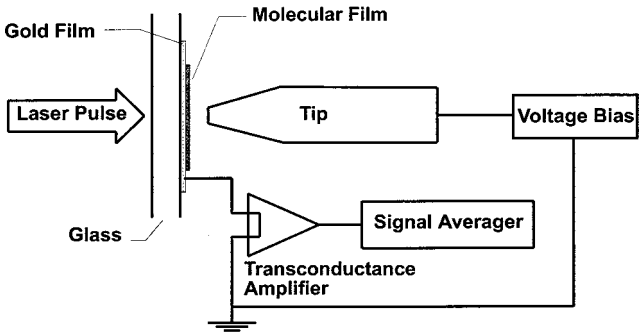

FIG. 1. Schematic of the experimental setup.

II. EXPERIMENTAL DETAILS

Figure 1 is a schematic of our experimental set-up. Our main light source is an argon-ion laser, running single-line at 488 nm, the output of which is gated with an acousto-optic modulator. The first-order-diffracted output of the modulator has a maximum power of about 100 mW and typically, we use pulses of 400 μs duration at 1 kHz repetition rate. Light is transmitted to the STM through approximately 5 m of single-mode optical fiber and finally focused through the glass substrate and semitransparent metal film into the junction using a microscope objective lens. The STM is of somewhat unusual design, but for present purposes the features to note are these: All motion control components are external to the small stainless steel and ceramic sample chamber, which contains no plastics, epoxies, etc. and which can provide a moderately high vacuum environment when needed. We find that the general features of our results are fairly insensitive to the ambient conditions in the experimental chamber. Exposure to air will change the conductivity of molecular films, but may have a lesser effect on the states that we observe. However, when film preparation is improved and experimental procedures are better understood changes in the transients with ambient conditions should be measurable. At present, for ease of sample and tip interchange, we have operated in air. The junction is formed between a scannable platinum-iridium or gold tip and a semitransparent gold-chromium film, typically 30 nm thick, evaporated onto an optical window. As we shall discuss below, it is essential that the tip act as a counterelectrode. For this reason, it is flattened by slowly pushing it against a clean glass dummy substrate. The fine control of this process afforded by piezotubes enables any flattened area to be created up to approximately 200 μm diameter. Typical tip flats are comparable in size to the focused beam spot size of approximately 20 μm (see Fig. 2).

The semitransparent film is connected to ground through a transimpedance amplifier and a bias voltage can be applied between the tip and ground. As the effects we are studying are transient, we use light pulses of suitable length and repetition rate and signal average the current response of the transimpedance amplifier. In measuring time-dependent small currents in an STM, one is greatly limited by the signal bandwidth of the transimpedance amplifier, but with a repetitive signal it is possible to trade some gain for bandwidth and average to obtain a usable signal. We have also experimented with charge amplifiers, which have considerably greater bandwidth, but which are less convenient in monitoring, for instance, the approach of the tip to a sample.

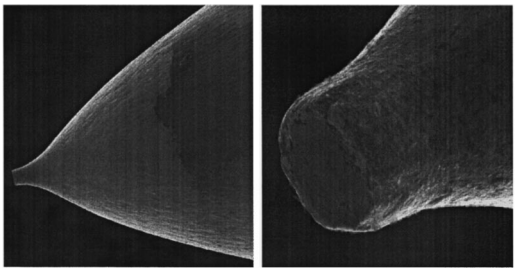

FIG. 2. Scanning electron microscope image of the side and end of a platinum tip with 10-μm-diam flat.

At present, we are concerned with establishing the general features of the transient behavior and have chosen to use several types of molecular films that can be prepared relatively easily. These are as follows: a mixture of high molecular weight hydrocarbons (HMWHC) obtained by spin coating from a solution of Apiezon ''*N*''[11] vacuum grease in trichloroethylene; spin-coated polymethylmethacrylate (PMMA), using chlorobenzene as solvent; self-assembled octadecanethiol; and anthracene, deposited from solution by evaporation of the solvent, toluene.

Under a small applied bias voltage (typically 100 mV) we find that it is relatively easy to approach the surface of a molecular film with the STM tip and detect current flow through the film at our standard setting level of 20 pA. We note that this surface cannot, however, be very precisely located and if the feedback loop is engaged, the tip position fluctuates up and down by approximately 1 nm. This is certainly not surprising, as conduction through the film is not by a vacuum tunneling mechanism. Having found the molecular film surface, the tip is retracted a distance of a few tens of nanometers, at which the steady-state current is zero. (If the tip is too close to the film, thermal expansion will cause it to make contact with the molecular film during the illumination period.) When light pulses are focused into the junction we observe transients of the type shown in Fig. 3(a), which were obtained with a HMWHC film about 30 nm thick under zero bias. Our method for measuring the thickness of the molecular films consists in establishing the location of the molecular film surface and then extending the piezo until the metal film surface is reached, at which point the current rises sharply. This procedure is used only at the end of a series of runs because it obviously damages the molecular film, and also transfers a significant amount of the film onto the tip, thereby confusing our interpretation of the transients. It is likely that surface forces distort the thickness measurement significantly. Ellipsometry would probably be a superior technique for future work and is compatible with our experimental arrangement.

III. ORIGINS AND CHARACTERISTICS OF THE TRANSIENT BEHAVIOR

A. Nature of trap states in molecular films

Insulators are often discussed in terms of a one-electron energy band picture, with valence and conduction bands, although this picture is of limited applicability. In the case of

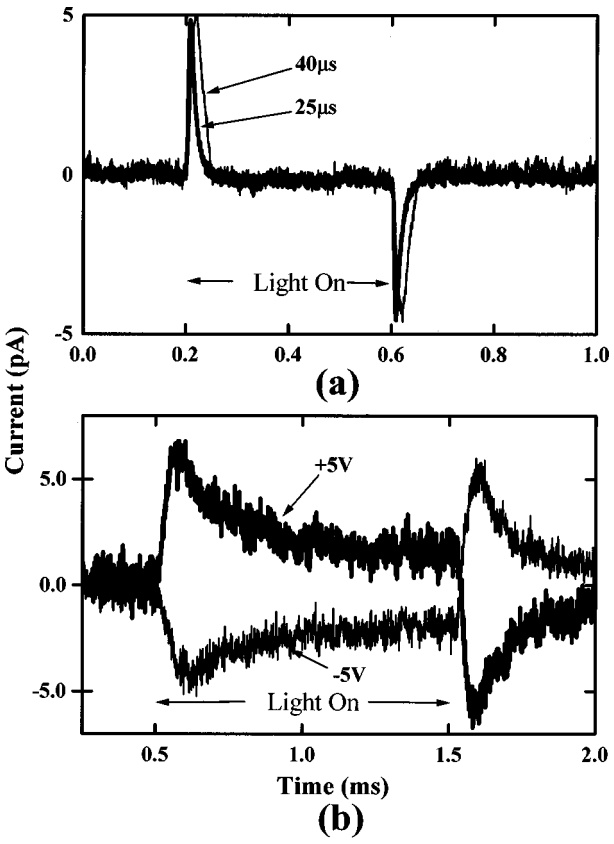

FIG. 3. (a) Transient current responses at zero bias for a 30-nm-thick HMWHC film obtained with different amplifier rise-time values, 40 μs (light line) and 25 μs (heavy line). (b) Thermal expansion responses at finite biases, $+5$ V (heavy line) and -5 V (light line).

molecular films, this picture can still be useful, but the bands should more properly be associated with the highest occupied and lowest unoccupied molecular orbitals. The band description recognizes that charge transport does occur through the orbitals. It is hoped that eventually our technique may provide useful information on this transport through orbitals, but for the present purposes we are focusing on the mechanism by which the insulator trap states finally communicate with the metal electrode. In many real insulators, the presence of disorder will smear the band edges, leading to one type of gap state. However, in the films we are using, which contain molecules bonded by van der Waals interactions, the smearing should be small and will therefore contribute few gap states.

Two other types of gap states might be expected to be important. First, there may be surface states. It has been suggested[12] that contact electrification of insulators involves only surface states. This suggestion is based on the observation that contact electrification is quite rapid. As the band gap of an insulator in contact with a metal would be expected to be aligned roughly symmetrically about E_F (in the absence of current flow) only surface states and mid-gap bulk states would be relevant. The existence of surface or interface states due to dangling bonds, defects, impurities etc. is certainly to be expected. However, the density of these at midgap is probably not large (see the various arguments in Ref. 1). On the other hand, bulk localized states at mid-gap are predicted for polymers by a well-known model.[13] However, if the bulk states are localized then charge cannot penetrate the insulator through them. Nonetheless, we observe that a nonzero current results when the tip makes contact with, or reaches tunneling distance from, the molecular films under any bias. This observation is consistent with many other experiments, for instance those of Ref. 10. Current flows through films much thicker than a tunneling distance and it therefore appears that charge does indeed penetrate into molecular films through bulk states near midgap. We postulate that the states are essentially those of Ref. 13, but that either the localization length is not small or that the density of the states is high enough that there is an effective trap band with considerable transport via tunneling, which can also be photoassisted in the present experiments.

B. Charge transport mechanisms under illumination

Light incident on the junction will excite electrons in both the metal electrodes and in the insulating film. In the case of the tip electrode, there are no accessible states near the metal and the electron energies are less than the work function, so we will assume that there is no electron displacement from the tip. On the other hand, electrons can be excited in the metal film and enter states in the insulating film via photoassisted tunneling. Electrons occupying traps in the insulating film can also be excited and move into the metal film. We might expect from the model of Ref. 13 that the relevant traps lie near the center of the band gap. As previously mentioned, the model we shall be using does not explicitly address transport between trap states in the molecular film, so we are assuming that transfer of charge between the metal film and the traps is the dominant transport process. While this may not be a good assumption for some molecular films, even some of the ones we have studied so far, this simplified picture provides a framework in which we can derive the expected transient behavior.

Under illumination, there are four current components: (1) the photocurrent from the metal film to the insulator; (2) the photocurrent from the insulator to the metal film; (3) the ''dark'' current from the metal film to the insulator previously described as the basic process of contact electrification—the migration of conduction electrons from the metal film to traps in the insulator; (4) the current from occupied states in the insulator above E_F to available conduction states in the metal film. In the transient condition at the beginning of the light pulse, there will generally be a net current into or out of the insulator. The displacement of electron charge in the insulator will cause a change in the charge induced on the electrodes. Now, in the absence of the tip, the induced charge on the substrate would, ignoring edge effects, remain equal and opposite to the electron charge and no current would flow through the transimpedance amplifier. However, if a counterelectrode (i.e. the tip) is present the induced charge is shared between the two electrodes. Displacement of electrons will cause a transfer of charge between the electrodes and current is registered by the amplifier. At the end of the light pulse there will be another transient as the traps return to equilibrium occupation. Again the electron displacement will cause a current, of the opposite sign to that of the beginning transient, to be registered by the amplifier.

The magnitude of the charge induced in the metal film due to electron motion in the insulating film depends on the spacing between the metal film and the tip and one can quite easily (and correctly) suppose that the fraction of the induced charge on each electrode is given by a simple ''lever rule.'' This result was originally justified in a general argument by Shockley[14] and later in a rigorous solution of the Poisson Equation by Fong and Kittel.[15] The magnitude of the charge transferred during a displacement of an electron between two trap positions is $\epsilon e(z_1-z_2)/L$, where ϵ is the dielectric constant of the insulating film, e the electronic charge, z_1 and z_2 distances of traps from the interface between the metal film and the insulating film, and L the distance between that interface and the tip. Note that this expression relates to any displacement of charge. It can describe equally well a displacement within the insulating film or across the interface between the metal film and the insulating film. In general, there will be many trapped electrons and so values z_{AV} and Δz_{AV} for the centroid of the trapped charge distribution and its displacement are appropriate.

In Fig. 4 the dependence of the transient magnitude on the displacement of the tip is plotted for a HMWHC film. Fitting the data with a reciprocal function $(d+d_0)^{-1}$ where d is the displacement of the tip from its initial position about 20 nm above the surface of the molecular film, gives a value $d_0 = 98$ nm, which is in fair agreement with the ultimately measured 70 nm film thickness.

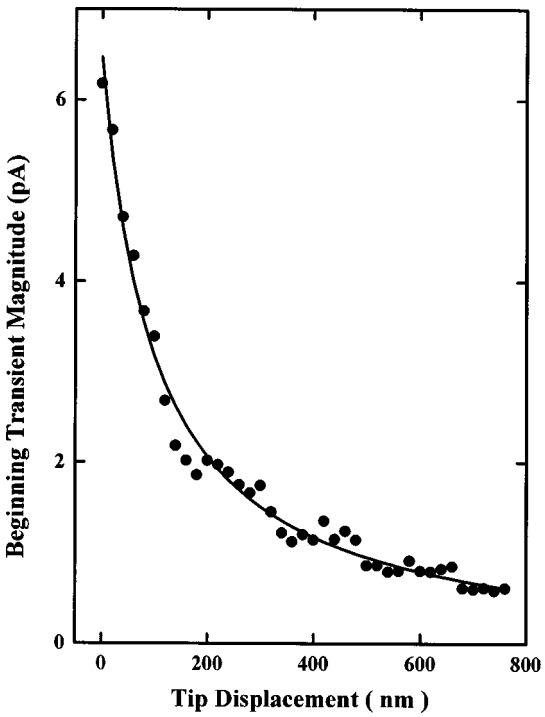

FIG. 4. Dependence of (beginning) transient magnitude on tip position. The zero value for tip position corresponds to the tip being about 20 nm away from the free surface of the insulating film. Fitting the data using the expected inverse relationship of transient magnitude to total tip-sample distance $d+d_0$ gives the solid line with $d_0=98$ nm. Note weak oscillations of period $\lambda/2$ due to weak Fabry-Perot behavior of the junction.

C. Modeling the transients

The transient response peaks shown in Fig. 3(a) are somewhat broadened by convolution with an instrumental response due to the finite bandwidth of the current amplifier, as evidenced by the difference between the responses measured with the different time constants. The ''beginning'' transient, corresponding to the beginning of the light pulse, in this instance is what we shall define as ''positive.'' The convention we are adopting is that a ''positive'' transient would correspond to electron motion toward the tip. Beginning transients of both polarities are seen, sometimes for nominally the same sample material. We shall discuss this later. To describe the transient behavior we will consider the model shown in Fig. 5. The trap states form a distribution of width E_T much less than both the insulator bandgap and the photon energy E_P. As mentioned, we believe that there is lateral transport within the trap distribution (as evidenced by the fact that the traps fill and refill from the metal), but we simplify the model by eliminating the lateral extent of the distribution. The electron motion between traps and metal then occurs only by photoassisted tunneling and potential driven (nonphotoassisted) tunneling. In this simplified model, if an electron is excited it is always displaced by the distance Δz_{AV} defined above. For a particular z_{AV} and L, calculation of the form of the current transients essentially reduces to solving a rate equation for the number current dN/dt, which must then be multiplied by a factor $\epsilon e \Delta z_{AV}/L$ to give the current.

We define rate constants P_I and P_M for photoassisted tunneling from the insulating film to the metal electrode and from the metal electrode to the insulating film, respectively. Similarly, rate constants for the potential driven processes are defined as T_I and T_M. The fact (to be discussed later) that the transients can be influenced by junction biasing suggests that E_F falls within the trap distribution. We shall make the approximation that E_P is considerably larger than the energy range of the trap band so we can consider photoassisted transition rates into or out of traps to be energy independent. Further, we shall assume that the density of states of the traps and of the metal are constant. As shown in Fig. 5 we will take a (generally bias-dependent) fraction $\alpha(V)$ of the total N traps to lie below E_F. In the fairly realistic situation in which traps are non-interacting we can separately consider the kinetics of traps above and below E_F. We will call the filled traps above and below E_F $N_>(t)$ and $N_<(t)$ respectively and now write two rate equations:

$$\frac{dN_>}{dt} = P_M\{(1-\alpha)N - N_>\} - T_I N_>, \tag{3.1}$$

$$\frac{dN_<}{dt} = -P_I N_< + T_M(\alpha N - N_<). \tag{3.2}$$

These equations can be solved and $N_>(t)$ and $N_<(t)$ summed to give the total number of filled traps at time t. For the beginning transient we obtain the following expression for the current:

$$I_B = [P_M(1-\alpha)\exp\{-(P_M+T_I)t\} - P_I\alpha\ \exp\{-(T_M+P_I)t\}]F, \tag{3.3}$$

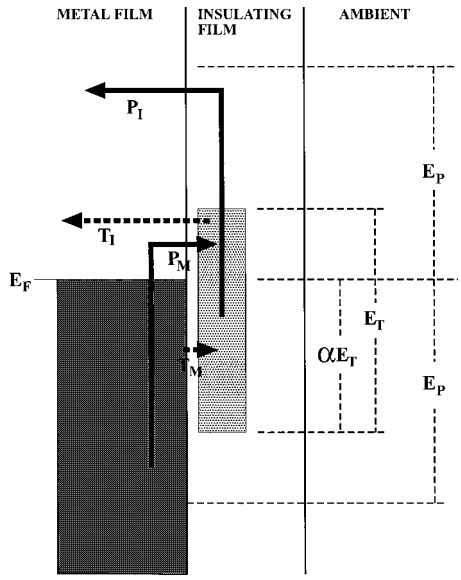

FIG. 5. Energy diagram for the midgap region of the interface between the metal film and the insulating film. Arrows indicate the four current components, which are labeled by the rate constants introduced in the text.

where we write F for the factor $\epsilon e \Delta z_{AV}/L$.

Similarly we obtain the following for the ending transient current:

$$I_E = \left[-\frac{P_M T_I (1-\alpha)}{P_M + T_I} \exp(-T_I t) + \frac{P_I T_M \alpha}{P_I + T_M} \exp(-T_M t) \right] F \tag{3.4}$$

At this point Eqs. (3.3) and (3.4) are merely descriptions of the transient behavior, because the four rate constants are unknown and no specific relationship between them is implied. However, one can see from the equations that the ending transient should generally have a longer time constant than the beginning transient. Further, integration of the currents for the two transients gives the equal and opposite charge required by charge conservation.

Often, the transients are only weakly bias dependent. In these cases, depending on whether the beginning transient is positive or negative, we postulate that the traps lie almost entirely above or below E_F, respectively. Equations (3.3) and (3.4) are now simplified. For the positive beginning transient case, $\alpha \approx 0$ and Eqs. (3.3) and (3.4) reduce to

$$I_B \approx [P_M \exp\{-(P_M + T_I)t\}] F \tag{3.5}$$

and

$$I_E \approx \left[\frac{P_M T_I}{P_M + T_I} \exp(-T_I t) \right] F. \tag{3.6}$$

Similarly, for the negative beginning transient case, $\alpha \approx 1$ and Eqs. (3.3) and (3.4) reduce to

$$I_B \approx [-P_I \exp\{-(T_M + P_I)t\}] F \tag{3.7}$$

and

$$I_E \approx \left[\frac{P_I T_M}{P_I + T_M} \exp(-T_M t) \right] F. \tag{3.8}$$

From Eqs. (3.5) and (3.7) we see that the initial value of the current in the model is either $P_M F$ for positive or $P_I F$ for negative beginning transients. In principle, as $P_M, P_I \rightarrow 1$ for very high light intensity we could obtain a saturation value for the quantity $F = \epsilon N \Delta z_{AV}/L$, which would be a lower bound to the number of traps, but in the absence of an independent measurement of Δz_{AV} we cannot determine the actual number of traps. (The discrepancy between the lower bound and the actual value will be very large when most of the traps are close to the metal substrate.) At the light intensities used in these experiments, we have not yet observed a significant deviation of the transient signal from direct proportionality to the light intensity and so we cannot extrapolate to saturation. This may, however, be achievable with different samples. (The maximum light intensity at the molecular film is about $2 MW/m^2$.) We note that the absence of saturation allows us to conclude that the trap occupation is low enough that there is no significant interaction between the traps.

D. Significance of bias dependence of the transients

While the observed current transients agree with the behavior of our model, it is still important to establish that our results truly are a consequence of trap kinetics, rather than other effects such as thermal expansion or thermoelectricity. Thermal expansion of the tip under laser illumination certainly can be observed. It was first discussed by Amer[16] in connection with measurements at tunneling distance. We do not operate with such small tip-sample distances and so the effect is much less significant. Nonetheless, it can sometimes be observed as a contributor to our transient response when using a finite bias. Figure 3(b) shows an extreme example of this, in which we have used bias voltages of ± 5 V in a junction with no insulating film present. As can be seen, there is no evidence of transients of the type shown in Fig. 3(a), which have time constants of less than 15 μs. Instead, we see much longer time constants of about 0.3 ms. Further, it is easily established that thermal expansion cannot be responsible for the Fig. 3(a) transients because these are observed even under zero applied bias.

It is quite possible that heating of the junction by the laser beam could give rise to a thermoelectric potential difference. However, this potential difference would presumably appear and disappear at the beginning and end of the illumination period with time constants comparable to those of the thermal expansion effect. The associated current transients would thus have even longer time constants, contrary to observation. Finally, we note that the mechanism that we have proposed in our model will cause charge to move ''against'' an applied field when the light is on, because the field primarily determines the value of the parameter α. This behavior is actually seen, as shown in Fig. 6. The film for which these data were obtained showed a somewhat larger bias dependence than a typical film, but all films display the effect. In order to obtain the greatest effect the tip must be held as close as possible to the molecular film surface, thereby maximizing the field strength within the film. Note that consider-

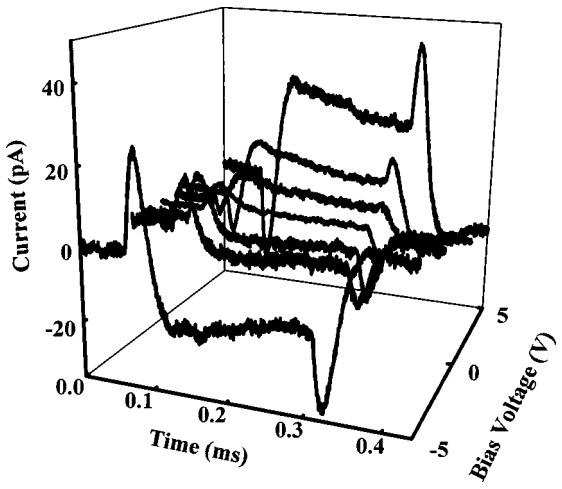

FIG. 6. Bias dependence of the current transients for an approximately 20-nm-thick HMWHC film.

able thermal expansion components, with typical long time constants, are seen at large biases. The simple fact that there is any bias dependence at all supports the picture that the traps constitute a distribution of a few eV in range spanning E_F. If there were a constant density of trap states throughout the insulator gap, then there should not be any bias dependence, but the finite width of the band results in a change in α.

IV. OTHER MOLECULAR FILMS

We have spent some time studying the HMWHC films because these are easy to prepare and display relatively large transients. Our intention, at this stage, is primarily to report the existence of the transients and establish some characteristics. However, we have also investigated the other films listed earlier in this work and can draw some interesting preliminary conclusions. Comparison of different films is complicated by the fact that the tips used have different flattened areas, which are not always measured with a scanning electron microscope. Ideally, the same tip would be used, but tips are sometimes damaged or must be discarded because they are suspected of becoming contaminated with the sample material. It should also be remembered that this transfer of material could change the transient behavior, leading to a complete reversal of the expected behavior in the event that most of the molecular film is located on the tip. We have tried to avoid this situation, but, except in the case of the self-assembled films, where there is a chemical bond of the thiol to the gold film, the molecular film can easily transfer if the tip is brought into contact with it. Following are some details of the transient behaviors observed with different molecular films.

A. PMMA

It appears to be the case that, whereas the HMWHC films have positive beginning transients, PMMA films have smaller (by about an order of magnitude for comparable light power and tip size), but negative beginning transients. [see Fig. 7(a)]. Note that the time constants for both transients are longer than those for the HMWHC films.

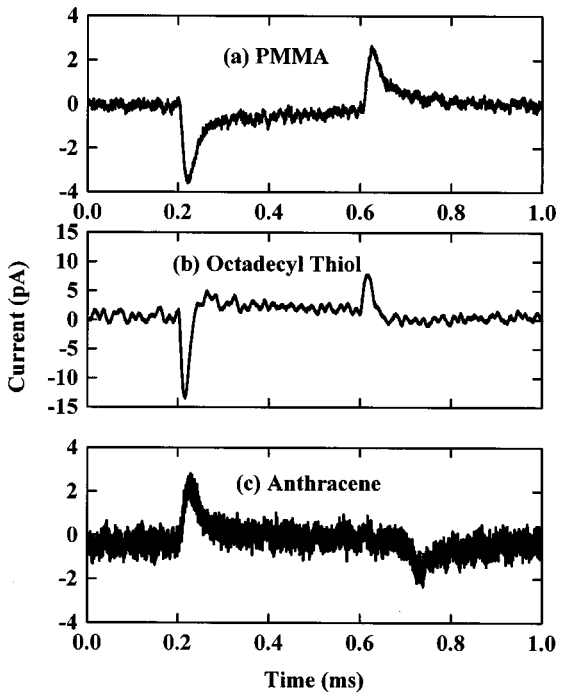

FIG. 7. Transient current responses at zero bias for various sample films: (a) PMMA film approximately 100 nm thick. (b) Octadecanethiol film 2.5 nm thick. (c) Anthracene film about 50 nm thick.

B. Self-assembled thiol

The self-assembled thiol films also have negative beginning transients that are somewhat smaller than the HMWHC transients. However, given that the thiol films are more than an order of magnitude thinner than all of the other films, the transients are comparatively large. This is perhaps consistent with an interfacial location of the trapped charge and also with the fact that only the thiol films form a chemical bond to the gold, leading to considerable charge transfer. Note in Fig. 7(b) that there are also small transient contributions of sign opposite to those of the main transients, but with much longer time constants.

C. Anthracene

Anthracene films have positive beginning transients that appear to be more than an order of magnitude smaller than those of the HMWHC films, as can be seen in Fig. 7(c). We tentatively interpret the different signs of the transients for the above films, in light of the argument presented earlier, as indicating what fraction of the traps lies above E_F. Only the transients of the HMWHC and anthracene films showed clear bias-dependence and so another tentative conclusion is that the trap distributions in these materials span E_F. While this sort of information can be obtained by varying the bias, much more information would result from variation of the wavelength of the light. So far, we have only used two wavelengths from an argon ion laser and a 100-nm range of longer wavelengths from a dye laser operating with Rhodamine 6G dye. No significant difference in the transient behavior was noted. In order to draw any useful conclusions wavelengths ranging over at least the visible spectrum should be used. This might seem straightforward, but it must

be borne in mind that the beam quality necessary for good coupling to the junction cannot easily be obtained in conjunction with adequate power. However, some well-collimated strong white light source and monochromator might be suitable with longer signal averaging.

V. CONCLUSIONS AND FUTURE PROSPECTS

The behavior of current transients observed in molecular films is consistent with photoassisted population and depopulation of states in the films. We have modeled this behavior using a band model of an insulator in which charge moves into and out of trap states near the center of the band gap. Such a simple model is useful for the purposes of this initial report of the transient behavior, but will need replacing with a more sophisticated treatment when improved measurements are made. For instance, transport through molecules can be discussed in terms of ''through bond'' and ''through space'' tunneling.[17] This and other approaches explicitly consider the bonding within molecular systems and the properties of states which we have somewhat simplistically considered as traps.

It should be possible to improve the transient measurement technique enough to make truly meaningful statements about the energetics of the charge in molecular films. This will require a sufficiently powerful tunable light source. We believe that there can also be considerable improvement of the time-resolution of the technique. The use of a charge amplifier, as has been mentioned, can be useful. However, to achieve a really significant improvement, it would be better to employ a method in which short-time information can be extracted from a low frequency measurement of displaced charge. Such a method, presently under development, uses optical pulse trains whose repetition rate can be modulated at low frequency. The intrinsic relaxation time for transient charge motion can be determined from the magnitude of the charge modulation.

ACKNOWLEDGMENT

This work was supported by the National Science Foundation, Contract No. NSF-DMR9401812.

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