White-light-induced disruption of nanoscale conducting filament in hafnia

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White-light-induced disruption of nanoscale conducting filament in hafnia

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Nanoscale conducting filament, which forms the basis of the HfO₂ resistive memory, is shown to exhibit a “negative photoconductivity” behavior, in that, electrical conduction through it can be disrupted upon white-light illumination. This behavior should be contrasted against the positive photoconductivity behavior commonly exhibited by oxides or perovskites having narrower bandgaps. The negative photoconductivity effect may be explained in terms of a photon-induced excitation of surrounding oxygen ions, which leads to migration and subsequent recombination with vacancies in the conducting filament. The finding suggests possible electrical-cum-optical applications for HfO₂-based devices, whose functionality is limited to-date by electrical stimulation.

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Visible-light-stimulated response of semiconductor devices^{1–7} may potentially expand the functionalities of electronics, which are presently limited to electrical stimulations. It was shown² that the current flow through the multilayer Pd/Al₂O₃/SiO₂/p-Si structure could be modulated by trapping, in the Al₂O₃, of electrons optically generated in the p-Si substrate. The finding provides a possible means for light-enabled information encoding in non-volatile memory applications. Light-stimulated resistive switching has been observed in structures such as the ZnO nanorod,³ BiFeO₃ thin film,⁴ TbMnO₃/Nb:SrTiO₃,⁵ ZnO/SrTiO₃,⁶ and TiO₂/α-Fe₂O₃⁷ heterojunctions. Although the actual response may vary with material characteristics and fabrication conditions, and the underlying mechanisms are still subjects of further investigation, these studies demonstrate that light sources may serve as an additional parameter for enhancing semiconductor device functionalities.

With the exception of Ref. 2, where the photoconductivity effect arises from electron-hole pair generation in the Si substrate, nearly all studies on light stimulation reported to-date have focused on photo-responsive oxide or perovskite materials having relatively small bandgaps (~3 eV or lesser).^{3–7} On the other hand, oxides such as SiO₂ and HfO₂, which are widely deployed in mainstream integrated-circuit manufacturing, have been excluded due to their much larger bandgaps (~6 eV or larger) which render them non-photo-responsive. In this study, we show that these wide-bandgap oxides can also be rendered photo-responsive by following an electrical-stress induced breakdown. In particular, it is shown that white light can either permanently or temporarily disrupt the nanoscale conducting filament in the HfO₂ dielectric. In recent years, dielectrics such as HfO₂ and Ta₂O₅ have been intensively studied as the active layer of resistive random access memory (RRAM) devices,^{8–10} by virtue of their compatibility with the mainstream integrated-circuit manufacturing technology. In the RRAM device, the change

between high and low resistance state is usually induced by applying an electric field, which modifies the oxide resistivity by repeatedly disrupting and reforming a nanoscale-width conducting filament through the oxide film separating two metal electrodes. Here, we reveal an intriguing observation that the conduction through a nanoscale conducting filament in the HfO₂ can be disruption by white light, an effect which we termed as the negative photoconductivity (NP) of the soft-breakdown HfO₂. In addition, the filament resistance can be controllably changed to desired values by varying the duration of a light exposure. These findings point to the possibility of direct implementation of light-enabled functionalities using HfO₂-based devices.

The measurements were performed at 300 K using a conductive atomic force microscope (C-AFM) in ultra-high vacuum (UHV) (~10^{−10} Torr), as depicted schematically in Fig. 1(a). The test sample comprises of a 4-nm thick HfO₂ layer, formed via the process of atomic-layer deposition on the TiN/Ti/p-Si substrate. Tetrakis (dimethylamino) hafnium was used as the metal precursor and H₂O vapor as the oxidizer. The growth temperature and pressure were 250 °C and

FIG. 1. (a) Schematic of the experimental setup used in this study. The probe of the ultra-high vacuum (UHV) conductive atomic force microscope is connected to a source/monitor unit of a semiconductor parameter analyzer. A voltage ramp applied to the probe is interrupted when an abrupt current increase is registered, indicating the formation of a conducting filament (cf) on the probe-contact site. A white LED lamp, placed at a quartz window of the UHV chamber, at a distance of ~30 cm from the test sample and at an angle of 45°, is used for the light exposure experiments. (b) White-light illumination of the TiN/Ti/p-Si sample shows an increase in current.

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0.1 Torr, respectively. No post-deposition annealing was carried out. A diamond-coated Si probe, connected to an external parameter analyzer, was used to form a conducting filament. A voltage-sweep was applied on the probe and the forming process interrupted when the increase in current reached a pre-set compliance limit. It should be clarified that the set-up comprising the uncapped HfO_2 sample with the C-AFM probe functioning as the top electrode is to enable the white light to reach the filament underneath the probe via its periphery, as the top metal electrode in the typical metal-insulator-metal (MIM) structure of a HfO_2 RRAM device impedes light transmission to filament(s) embedded in the oxide. Surface contamination of the sample during testing was avoided by the UHV environment, which is not a prerequisite for the observation of the NP behavior. In principle, a MIM structure that has an optically transparent top metal electrode should exhibit the NP behavior.

The white light source (of wavelengths ranging from ~ 400 to 700 nm) used in this study was a standard light-emitting diode (LED) lamp, positioned at a quartz window of the UHV chamber at a distance of ~ 30 cm from the sample. The angle of incidence was $\sim 45^\circ$. Measurement under light exposure was also performed on a bare TiN surface (i.e., a control sample without an active oxide layer). Fig. 1(b) shows a typical photoconductive response (i.e., current increase upon illumination) for this control sample.

Fig. 2(a) depicts the light-induced disruption of a nanoscale conducting filament in HfO_2 , as evidenced from the comparison to the measurement on the same sample performed with no light exposure. Following the formation of a conducting filament using a positive voltage ramp (Fig. 2(b), the filament current was monitored at a probe voltage of 1 V as a function of time. The relatively large resistance (~ 10 M Ω) of the filament may be attributed to its very small diameter of several nanometers (as revealed later in Fig. 4). The estimated filament resistivity is on the order of 10^{-2} $\Omega\cdot\text{m}$, corresponding to that of a semiconductor-like material. As can be seen in Fig. 2(a), the current decays slowly over the 2000-s time interval, in the absence of light exposure, as expected from post-electrical-stress relaxation. Re-measurement following the first 2000-s interval shows a constant current ($\sim 10^{-8}$ A), after the relaxation was completed.

FIG. 2. (a) Current versus time characteristics of a conducting filament: circle—measurement made immediately after forming in darkness throughout; square—re-measurement following the initial 2000-s period with a brief exposure to white-light as indicated. (b) Current-voltage curves: line—formation of the filament in (a); square—after the white-light exposure indicated in (a); triangle—electrical reset by an opposite-polarity voltage sweep; and circle—after electrical reset. Arrows indicate directions of voltage sweep. The current compliance limit (CCL) is 100 nA.

Interestingly, a significant decrease of the current, from $\sim 10^{-8}$ A to the measurement floor ($\sim 10^{-15}$ A) can be observed upon illumination with a white light, in contrast to the behavior shown in Fig. 1(b) for the control sample. After the light source is removed, the current recovers slightly to $\sim 10^{-14}$ A and remains at this level for the remaining duration of the measurement. A subsequent positive voltage sweep yields a current similar to the pre-forming level (Fig. 2(b), square), thus confirming that the conducting filament was disrupted during the illumination. Apparently, the white light has “reset” the conducting filament to a high resistance state. A subsequent transition back to a low resistance state, however, occurs at a lower positive voltage as compared to the initial forming process, indicating that the oxide region concerned was not fully restored to its pre-forming state. Following a typical bipolar (electrical) reset using a negative voltage sweep (triangle), a post-electrical-reset current-voltage (I - V) curve (circle) comparable to the post-light-reset I - V curve (square) is attained, indicating that the effect of light is similar to that of the electrical reset. By alternating between the light and electrical reset modes, similar post-reset I - V curves are obtained regardless of the type of the prior reset mode. The light-induced resistance reset revealed in Fig. 2(a) is also observed in ZrO_2 and SiO_2 samples.¹¹

It can be noted in Fig. 2(a) that the current decrease occurs after the sample was exposed to the light for ~ 100 s. To investigate this “delayed response,” the experiment was repeated on an untested location using two different light intensities. Initially, light of a lower intensity was applied to induce reset after forming. Then, the conducting filament was reformed by ramping up a positive voltage followed by the exposure to a light of a five times higher intensity. As can be seen in Fig. 3(a), the current decreases (following an initial no-exposure period of ~ 20 s) more gradually under the lower intensity light exposure, as compared to the higher intensity case where the current decreases spontaneously and more abruptly (showing a reset that occurred at a distinctively faster rate). In the case of the low-intensity light, it takes 52 s for the current to decrease to the measurement floor. As for the case of the high-intensity light, the time is

FIG. 3. (a) Dependency of the conducting filament reset speed on the light intensity. The initial reset to a high resistance state is performed using a light intensity of 1 mW/cm 2 , followed by a set operation to a low resistance state performed by applying a positive voltage sweep (no light exposure; note that this state is less resistive than the initially formed, pre-first-exposure state), followed by a much faster reset to a high resistance state carried out with the higher light intensity of 5 mW/cm 2 . (b) An intermediate resistance level (HRS1) can be achieved by limiting the duration of exposure at a given light intensity; LRS—an initial low resistance state; HRS1—an intermediate high resistance state after the exposure; and HRS2—a final high resistance state after the additional exposure.

FIG. 4. C-AFM current maps: (a) relatively uniform current distribution before forming; (b) a bright shade of ~ 4 nm in diameter denoting the conducting filament after forming (the peak current is > 1 nA); (c) reduced filament size and brightness (i.e., current) after a 30-s white-light illumination; and (d) near-complete filament disruption upon a further 300-s illumination. Inset: Remnants of the filament are visible only at a much reduced current scale.

9.3 s. This 5.6-time reduction corresponds well to the difference between the two light intensities. Clearly, light intensity strongly influences the speed at which the conducting filament is disrupted. The current evolution in the absence of illumination is also shown for comparison.

As the data in Fig. 3(a) show, the change in resistance in a specific time interval is determined by the amount of light energy that the filament received, indicating a possibility of achieving multiple resistance levels for a given filament. This is indeed observed in Fig. 3(b), where different levels of the conducting filament disruption can be controlled by changing the duration of light exposure. Upon illumination, the current is decreased from the low-resistance state (LRS) to a first high-resistance state (HRS1) in a 20-s interval. The current decrease is obviously arrested when the light illumination is removed, as is evident from the current plateau. When the illumination is resumed, the current is decreased to the next higher resistance state (HRS2). Clearly, by controlling the light exposure, one can generate multiple resistance values for the conducting filament. This result suggests the possibility of a light-enabled RRAM-resistance tuning approach, as an alternative to the electrical counterpart, for neuromorphic computing applications.¹² The electrical tuning method might be challenging because it requires the control of a set current compliance limit (for DC switching) or set/reset pulse timing (for AC switching).¹³

To further illustrate that the disruption of current conduction observed in Figs. 2 and 3 is a consequence of light induced “dissolution” of the filament, current maps of a selected region of the HfO_2 before and after forming, and after various stages of light exposure are compared in Fig. 4. Compared to uniform current map before forming (Fig. 4(a)), the presence of a distinct bright shade, of ~ 4 nm in diameter, after forming (Fig. 4(b)) is due to the generation of a conducting filament. Interestingly, after a 30-s exposure to white light, the level of brightness and size of the filament are drastically reduced (Fig. 4(c)), which correspond to a decrease in current. Upon further illumination, the filament became almost indistinguishable from the surrounding oxide region (Fig. 4(d)). The results confirm that white light can eradicate a conducting filament formed in the soft-breakdown HfO_2 .

The light induced resistance-reset phenomenon discussed above should be distinguished from the photo-related resistance switching effects reported in recent studies.^{1–7} It was reported by Yu and Wang¹ that the resistance between two points along a thin Ti film could be continuously varied between two extreme values when a laser beam was scanned between these points. As shown in Ungureanu *et al.*,² the current through the $\text{Pd}/\text{Al}_2\text{O}_3/\text{SiO}_2/\text{p-Si}$ device biased in inversion was modulated by injecting optically generated minority carriers (electrons) in the p-Si substrate and trapping these electrons in the Al_2O_3 layer. Park *et al.*³ found that the resistance of a ZnO nanorod RRAM device was switched to a lower value at a smaller voltage under light exposure. After the light exposure, however, the resistance would transit towards a higher value at a rate determined by the decaying persistent photoconductivity (PPC) effect.¹⁴ A significant PPC effect in the $\text{ZnO}/\text{SrTiO}_3$ heterojunction obscured the electrically induced resistance switching of this device following light exposure.⁶ In heterojunction structures, the PPC effect has been ascribed to the trapping of charges at interface defects, which modulate the junction barrier controlling current flow.^{4–7} A common feature in the abovementioned studies is that the test-sample resistance is modulated by a significant photoconductivity effect. In contrast, the HfO_2 , ZrO_2 , and SiO_2 are non-photo-responsive materials due to their relatively large bandgaps. In contrast to photo-responsive oxides, white light has no impact on the I - V curve measured during the forming/set steps performed on HfO_2 . However, once the conducting filament is formed, a unique feature of the light-induced filament disruption (i.e., a “negative photoconductivity effect”) is observed in this study.

A possible explanation for the light-induced resistance reset phenomenon in HfO_2 is illustrated in Fig. 5, based on the oxygen migration model.^{13,15,16} The proposed explanation is guided by two recent findings: (1) observation of an oxygen-depleted filament via atomic-scale electron-energy loss spectroscopy;¹⁷ and (2) observation, by simulation, of oxygen ions released from the filament site during formation, in the vicinity of the filament.¹⁸ Fig. 5(a) depicts the formation of an oxygen-deficient conducting filament through the HfO_2 layer when a voltage sweep is applied via the C-AFM probe. Electron transport through the hafnia film induces Hf-O bond breakage and out-diffusion (assisted by the temperature and electrical field distributions) of the released negatively charged oxygen ions, resulting in the formation of an oxygen-deficient (metal-rich) conducting filament. As the forming process is interrupted when the current reaches a compliance limit, some of the released oxygen ions remain in the interstitial positions in the vicinity of the filament¹⁸ and may be subsequently driven back to the filament by the applied electrical field and temperature gradient during the electrically controlled reset.¹³ The diffusion of interstitial oxygen ions entails the overcoming of an energy barrier of 0.3–0.6 eV.¹⁹ At a low voltage (below the electrical reset voltage), the diffusion rate would thus be negligibly small. It is suggested that a white light illumination generates a photon-induced excitation of the oxygen ions over the diffusion energy barrier, hence accelerating their diffusion towards the vacancy-rich filament, as illustrated in Fig. 5(b). The ion excitation proceeds more efficiently under a higher

FIG. 5. Schematic illustration of the proposed mechanism for the light-induced resistance reset. (a) Formation of a conducting filament comprising oxygen vacancies, with the released oxygen ions populating interstitial sites in the filament vicinity. (b) Filament disruption by recombination between filament vacancies and photo-excited migrating oxygen ions (arrows) from the vicinity. (c) Filament formed at a higher current compliance has fewer interstitial oxygen in its vicinity due to increased lateral propagation of the released oxygen ions. (d) Partial filament disruption due to limited supply of photo-excited migrating oxygen ions, including weakly bonded lattice oxygen in the filament vicinity. Additional vacancies generated lead to oxygen back-diffusion (arrows) and filament reformation after light exposure.

light intensity, thus enhancing their migration back to the vacancy sites, as manifested by a faster resistance reset rate in Fig. 3(a).

The effect of the conducting filament dimension as determined by the current compliance limit (CCL) used during the DC forming process on light-induced reset is also studied and the results are shown in Fig. 6. When the conducting filament is formed at a low CCL (of 10 nA), the light-induced reset state is relatively stable (i.e., after the light is turned off, the current remains constant at a low level throughout the entire duration of the observation time). On the other hand, when the conducting filament is reformed at a higher CCL of 100 nA, the light induced reset is less complete and unstable (i.e., the

FIG. 6. Influence of the CCL used to form the conducting filament on subsequent light induced reset to a high resistance state. Light of a given intensity and exposure duration (10 s) results in a complete reset or disruption of the filament formed with a lower CCL (a). On the other hand, partial reset is evident for the filament formed at a higher CCL, as can be seen from the much higher current at the end of illumination. Moreover, a post-exposure gradual recovery of the current towards the level prior to illumination can be observed.

current after illumination is much higher than the case of low CCL and can be seen to gradually increase back to the pre-set level). Similar results are obtained on the $\text{ZrO}_2/\text{TiN}/\text{Ti}/\text{p-Si}$ sample.¹¹ These observations are consistent with the model discussed in Fig. 5. When a lower CCL is used during DC forming, fewer oxygen ions are released due to a smaller required filament cross-section. Since the process involved is shorter (terminated once the low CCL is reached), a larger portion of the released oxygen ions populate interstitial sites in the immediate vicinity of the filament (Fig. 5(a)). These interstitial oxygen ions may then recombine with the filament vacancies during light-driven reset (Fig. 5(b)). On the other hand, a DC forming process using a larger CCL releases more oxygen ions (due to a larger filament cross-section) and also enables a larger share of the released ions to propagate further away from the filament as a result of the greater Joule heating and longer process involved (Fig. 5(c)). Then, during a subsequent light exposure, a smaller share of the light-activated interstitial oxygen ions distributed throughout the dielectric volume may potentially reach the filament, resulting in only partial filament disruption. As the recombination of filament vacancies may also involve light-stimulated migration of weakly bonded lattice oxygen ions in the filament vicinity towards the filament vacancies, the resultant vacancies formed would in turn trigger the back-diffusion and reformation of filament vacancies (Fig. 5(d)) after the light was turned off. This could explain the gradual recovery of the current towards the pre-illumination level. It should be noted that in Fig. 2, the filament was also formed at a CCL of 100 nA but a stable light-induced reset was achieved (i.e., no increase of current after removal of illumination). The better reset stability may be attributed to the much longer light-exposure period (200 s) as compared to that of Fig. 6(b) (10 s).

In summary, we have shown that the resistance of a nanoscale conducting filament (formed using voltage sweep under a certain current compliance limit) in the HfO_2 dielectric can be increased by exposing the dielectric film to white light. Resistance values can be modulated by varying the light intensity and exposure duration. The light-induced resistance increase in the reset process is ascribed to a conducting filament disruption caused by a recombination of the oxygen vacancies there with oxygen ions, whose transport from the surrounding interstitial locations is activated by the white light. These findings point to a possibility of extending the application range of HfO_2 -based devices to the optical domain, e.g., imaging sensors and photodetectors with built-in non-volatile memory capability.

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