Model for petahertz optical memory based on a manipulation of the optical-field-induced current in dielectrics

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Abstract
A new physical insight into the ultrafast information communication can be gained from the reversible and robust petahertz (PHz, $10^{15}$ Hz) current induced by a strong few-cycle optical waveform in large band-gap dielectrics. We explore an asymmetric conduction of the petahertz current using a heterojunction of low-hole-mass and low-electron-mass dielectrics and devise various functionalities enabling the petahertz signal processing, like diode, switch, and diode transistor. We then propose a model of one-bit optical nonvolatile random-access memory (RAM) by assembling those functionalities and demonstrate its petahertz operation. Further, we suggest the scalability up to a four-bit data manipulation based on the $2 \times 2$ array of four one-bit RAM elements.

1. Introduction
An advancement of the high-intensity subfemtosecond laser source has given access to the extreme ultrafast and nonlinear phenomena of matter [1]. For instance, isolated and intense subfemtosecond laser pulses have provided massive real-time insights to the quantum process activated by the photoexcited electron dynamics of atoms, molecules, and solids on a time scale of 100 attoseconds (as)–1 femtosecond (fs) [2–19]. Recently, in particular, the subfemtosecond electron motion driven by the electric field oscillation of light opens the way to dramatically boost the working speed of electronics, which extends the clock rate of electronic signal processing up to the petahertz (PHz, $10^{15}$ Hz) domain.

Delicate light-wave-controlled electron dynamics observed in diverse nanostructures reveal the wave nature of light in the context of extreme ultrafast quantum mechanics [13–16]. Schiffirin et al [20] have experimentally demonstrated that a strong few-cycle laser pulse induces the electric current and macroscopic charge separation in SiO$_2$, a dielectric whose band gap $E_g$ is as large as ~9 eV, without optical breakdown. This is a remarkable discovery in that such reversible and robust current could provide a great potential for the future light-wave electronics enabling the processing of petahertz (PHz, $10^{15}$ Hz) electric signals. Recently, Mashiko et al [21] have presented the optical driving of carrier dynamics with a 1.16 PHz bandwidth in a wide-band-gap (~3.35 eV) semiconductor, GaN. Further, stimulated theoretical investigations on optical-field-induced currents in dielectrics have proceeded based on the independent particle models [22, 23], adiabatic band response [24], and first-principles time-dependent density functional theory [25].

An open question at the present stage will be whether the optical-field-induced current can be manipulated and made directly accessible to the signal processing [26]. One of the first meaningful attempts was a proposal of the petahertz diode for rectifying the optical-field-induced current [27], which is given by a heterojunction of two dielectrics with different polarities, i.e., low-hole-mass (LHM; $m_h^* < m_e^*$) dielectric with a large hole mobility and low-electron-mass (LEM; $m_e^* < m_h^*$) dielectric with a large electron mobility (figure 1). $m_h^*$ and $m_e^*$ represent the effective hole and electron masses. Those dielectric heterojunction devices may be key ingredients for the signal processing ensuring the petahertz manipulation.

Ultrafast optical memory is a challenge critical for an advancement of the information processing. In this letter, we propose a model for the petahertz optical memory based on a manipulation of the optical-field-induced current in dielectrics. A basic petahertz device made of the LHM-LEM or LEM-LHM
heterojunction is explored to have various functionalities like diode, switch, and diode transistor in an optoelectronic circuitry. Assembling those functionalities, we devise a prototype of one-bit optical nonvolatile random-access memory (RAM) and demonstrate its extreme ultrafast operation working at the petahertz control speed. We also discuss the feasibility of an integration of the one-bit RAM element in terms of the $2 \times 2$ elemental array up to the four-bit memory, which sets a viable milestone for an incorporation of the subfemtosecond electron dynamics into the ultrafast processing and communication.

2. Results

We consider a single heterojunction made of LHM and LEM dielectrics of figure 1(a) or (b), a basic petahertz device. Under a strong few-cycle optical pulse, the time-dependent macroscopic current can be studied by introducing a model Hamiltonian $\mathcal{H}$,

$$\mathcal{H} = \mathcal{H}_{\text{LHM}} + \mathcal{H}_{\text{LEM}} + \mathcal{H}_t.$$  \hspace{1cm} (1)

In equation (1), $\mathcal{H}_{\text{LHM}}$ and $\mathcal{H}_{\text{LEM}}$ are Hamiltonians introduced to describe dielectrics with the definite polarity corresponding to LHM and LEM as shown in figure 1(c), respectively, each of which is given by $\mathcal{H}_m + \mathcal{H}_t(\tau)$. $\mathcal{H}_m$ is a material term and $\mathcal{H}_t(\tau)$ a time-dependent optical pumping term. $\mathcal{H}_m$ is taken as a standard one-dimensional (along the direction of $\mathbf{x}$) tight-binding Hamiltonian, i.e.,

$$\mathcal{H}_m = \sum_k \varepsilon_k c_k^\dagger c_k + \sum_{\langle k, k' \rangle} \varepsilon_{k-k'} d_k^\dagger d_{k'},$$

where $c_k$ ($c_{k'}$) and $d_k$ ($d_{k'}$) are the electron and hole operators with band energies of $\varepsilon_k = \varepsilon_v + (\Delta_v/2) \cos(ka)$ and $\varepsilon_{k-k'} = \varepsilon_v + (\Delta_v/2) \cos(ka)$ with the lattice constant $a$, respectively. The indices of $c$ and $d$ denote the conduction (electron) and valence (hole) bands. Depending on the desired functionality of the heterojunction device, $\mathcal{H}_t(\tau)$ could be written as optical transitions due to $A_1(\tau)$ or $A_{-1}(\tau)$, or both, i.e.,

$$A_j(\tau) = A_1 \exp(-\tau^2/\tau_W^2) \cos(\omega \tau + \delta) \hat{x} = A_j(\tau) \hat{x} \quad \text{and} \quad A_{-j}(\tau) = A_1 \exp(-\tau^2/\tau_W^2) \cos(\omega \tau + \delta) \hat{y} = A_{-j}(\tau) \hat{y},$$

for the laser parameters, we take $\tau_W = 4$ fs and $\omega = 1.7$ eV and the beam spot size to be comparable to the lateral dimension of the device. $A_j(\tau)$ contributes to the interband transition through $t_j' = i \langle c | P_j | v \rangle$ and $t_{-j}' = i \langle v | P_j | c \rangle$, where $|c\rangle$ and $|v\rangle$ are the $j$-centered electron and hole states, respectively. In contrast, both $A_1(\tau)$ and $A_{-1}(\tau)$ additively contribute to the intraband transition with $d_j = i \langle c | P_j | v \rangle$ and $d_{-j} = i \langle v | P_j | c \rangle$. Thus, $\mathcal{H}_m + \mathcal{H}_t(\tau)$ can be written as

$$\mathcal{H}_m + \mathcal{H}_t(\tau) = \sum_k \{ \varepsilon_k + 2t_j A_j(\tau) \sin(ka) \} c_k^\dagger c_k + \sum_k \{ \varepsilon_{k-k'} + 2t_{-j} A_{-j}(\tau) \sin(ka) \} d_k^\dagger d_{k'},$$

$$+ \{ A_j(\tau) d_j + A_{-j}(\tau) d_{-j} \} \sum_k (c_k^\dagger d_k^\dagger + d_{-k}^\dagger c_k).$$  \hspace{1cm} (2)

$\mathcal{H}_t$ describes the tunneling across the LHM-LEM heterojunction in the form $\mathcal{H}_t = V_{ce} + V_{ve} + V_{ce} + V_{ve}$, where $V_{ce} = T_{ce} \sum_{j,k,k'} (c_j^\dagger c_{k'} \Delta_{j,k'} \cos(ka) + \text{h.c.})$ and $V_{ve} = T_{ve} \sum_{j,k,k'} (c_j^\dagger c_{k'} \Delta_{j,k'} \cos(ka) + \text{h.c.})$ are given and $T_{ve}$ and $T_{ve}$ are defined similarly. For the present model calculation, we ideally arrange the extreme LHM and LEM dielectrics symmetrically. That is, the hole (electron) band of the former and the electron (hole) band of the latter...
In the end of calculation, putting \( T_{00} = T_{0v} = 340 \text{ meV} \) and \( T_{cv} = T_{vv} = 68 \text{ meV} \),

Dynamics are initiated by the optical pumping and described by the time-dependent Schrödinger equation

\[
\left( i \partial / \partial \tau \right) |\Psi(\tau)\rangle = \mathcal{H}_T |\Psi(\tau)\rangle,
\]

Explicit dynamical responses under several geometries of optical pumping are investigated up to the second order of \( \mathcal{H}_T \). \( |\Psi(\tau)\rangle \) is the quantum state that describes an entire heterojunction, given in the following:

\[
|\Psi(\tau)\rangle = C(\tau) |0, 0\rangle_{\text{LHM}} |0, 0\rangle_{\text{LEM}} + \sum_k C_k^p(\tau) |k, -k\rangle_{\text{LHM}} |0, 0\rangle_{\text{LEM}} + \sum_k C_k^p(\tau) |0, 0\rangle_{\text{LHM}} |k, -k\rangle_{\text{LEM}} + \sum_{kk'} C_{kk'}^v(\tau) |k, 0\rangle_{\text{LHM}} |k', 0\rangle_{\text{LEM}} + \sum_{kk'} C_{kk'}^v(\tau) |0, k\rangle_{\text{LHM}} |0, k'\rangle_{\text{LEM}},
\]

where \( |k, k'\rangle_{\text{LHM}} \) stands for \( c_{\text{LHM},k}^\dagger |0, 0\rangle_{\text{LHM}} \) on the LHM side and \( |k, k'\rangle_{\text{LEM}} \) on the LEM side similarly. One can then attain the electron and hole occupations in the LHM side,

\[
n_{\text{LHM}}(\lambda, \tau) = \sum_k |\langle \Psi(\tau)|c_{\text{LHM},k}^\dagger c_{\text{LHM},k}|\Psi(\tau)\rangle|^2
\]

and \( n_{\text{LHM}}(\lambda, \tau) = \sum_k |\langle \Psi(\tau)|d_{\text{LHM},k}^\dagger d_{\text{LHM},k}|\Psi(\tau)\rangle|^2 \),

where \( \lambda \) is simply a control parameter replacing \( T_{0v} \rightarrow \lambda T_{0v}, T_{00} \rightarrow \lambda T_{00}, T_{vv} \rightarrow \lambda T_{vv}, \) and \( T_{cv} \rightarrow \lambda T_{cv} \). In the limit of \( \lambda \rightarrow 0 \), one can then obtain

\[
n_{\text{LHM}}(\lambda, \tau) = n_{\text{LHM}}(0, \tau) + \lambda^2 N_{\text{LHM}}(\tau) + \ldots,
\]

\[
n_{\text{LHM}}(\lambda, \tau) = n_{\text{LHM}}(0, \tau) + \lambda^2 N_{\text{LHM}}(\tau) + \ldots.
\]

In the end of calculation, putting \( \lambda = 1 \), we guarantee that \( N_{\text{LHM}/\text{LEM}}(\tau) \) and \( N_{\text{LHM}/\text{LEM}}(\tau) \) are band occupations relevant to the lowest-order tunneling between the two dielectrics. Now the current across the heterojunction can be simply given by

\[
J(\tau) = \frac{\partial}{\partial \tau} \mathcal{P}(\tau) = -\frac{\partial}{\partial \tau} \left( N_{\text{LHM}}(\tau) - N_{\text{LHM}}(\tau) \right).
\]

A single in-plane pulse \( A_x(\tau) \) produces the rectified current from LHM to LEM dielectric (figures 2(a) and (b)), where the basic device plays a diode function [27]. The periodically oscillating population (gray line of

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For the LHM dielectric, we take \( \Delta \omega = -6 \text{ eV}, \Delta \omega = -6.5 \text{ eV}, \omega = 4 \text{ eV}, \omega = 7 \text{ eV} \) (leading to an energy gap of 7.75 eV), \( \epsilon_0/\omega = 0.15 \text{ Å}, \epsilon_0/\omega = 1.7 \text{ Å}, \) and \( d_0/\omega = d_0/\omega = 1.5 \text{ Å} \). The hole and electron parameters are exactly reversed for the LEM dielectric.

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\(^{1}\) For the LHM dielectric, we take \( \Delta \omega = -6 \text{ eV}, \Delta \omega = -6.5 \text{ eV}, \omega = 4 \text{ eV}, \omega = 7 \text{ eV} \) (leading to an energy gap of 7.75 eV), \( \epsilon_0/\omega = 0.15 \text{ Å}, \epsilon_0/\omega = 1.7 \text{ Å}, \) and \( d_0/\omega = d_0/\omega = 1.5 \text{ Å} \). The hole and electron parameters are exactly reversed for the LEM dielectric.
dielectrics can hardly make a directional charge transfer, can be understood through the optical interference induced by a complete overlap of the two pulses [28]. Consequently, we witness that the double-pulse optical pumping not only rectifies the optical-field-induced current but also amplifies ($\delta = 0$) or suppresses ($\delta = \pi$) the current and therefore note that the basic device operates as a diode transistor in figure 2(c).

Meanwhile, a single out-of-plane pulse $A_\perp (\tau)$ creates a role of switch to flow or stop the current under a constant bias electric field across the device (figure 3). At $E_{\text{bias}} \approx 0$, carriers excited by $A_\perp (\tau)$ in both sides of the semiconductor $-\Delta\tau$ are assumed. Hence, $A_\perp (\tau)$ induces the insulator–semimetal transition in the dielectric device within a quarter optical cycle and $E_{\text{bias}}$ enables a directional conduction of the sub femtosecond current. Recently, it has been reported that CaF$_2$ undergoes the transient metalization through a change of conductivity $\Delta \sigma \sim 10^{10} \text{ S m}^{-1}$ at the out-of-plane injection pumping [29].

We now put together two basic devices (i.e., LEM-LHM and LHM-LEM devices) and a parallel capacitor into a one-bit RAM (figure 4(a)) and examine the ultrafast dynamics of memory operations at the petahertz control speed. Figure 4(b) shows that an application of the in-plane pulse $A_\parallel (\tau)$ (i.e., write pulse) to LHM-LEM device lets the memory turned on by changing the charge state in a capacitor from ‘0’ to ‘1’. This clearly indicates the ultrafast operation of writing a one-bit information onto the memory. If necessary, the geometry of figure 2(c) may be adopted to amplify or suppress $Q_\perp (\tau)$ to the capacitor. Incorporating the optical-field-induced current $J (\tau)$ and the transient dc resistance $R(\tau)$ across the junction, we can explore the charge $Q_\perp (\tau)$ from $J (\tau) R(\tau) + \frac{dQ_\perp}{dt}[R_{\text{wire}} + R(\tau)] + \frac{1}{C_Q} Q_\perp = 0$. $R(\tau)$ would then be assumed to be $\propto 1/J_{\text{dc}}(\tau)$ with a parametrized minimum value of $R_{\text{min}}$ where $J_{\text{dc}}(\tau)$ is proportional to the optically excited carrier density $n(\tau)$ (gray line of figure 2(b)) in the geometry of figure 4(b). $R_{\text{wire}}$ is the wire resistance. Next, an application of the out-of-plane pulse (i.e., read pulse) to LHM-LEM device makes it possible to check the charge storage by inducing a small charge flow from the capacitor, which facilitates the read process by $\frac{dQ_\perp}{dt}[R_{\text{wire}} + R(\tau)] + \frac{1}{C_Q} Q_\perp = 0$. $R(\tau)$ is reflected in the insulator–semimetal transition depending on $E_\perp$ (figure 3(b)) and $E_{\text{bias}}$. In addition, $\propto 1/Q_\perp(\tau) \propto 1/E_{\text{bias}}$. Let us note that figure 4(c) manifests the operation of reading a one-bit information stored in the memory. The erase process for removing all the stored information and refreshing the memory is basically the same as the read process. That is, a strong out-of-plane pulse (i.e., erase pulse) would make all the charges flow away from the capacitor and then exhausted (figure 4(d)).

For both the write and read (also erase) processes, the condition of $R_{\text{wire}} \ll R_{\text{min}}$ is required for a proper performance as shown in figure 5. Otherwise, the write and read processes would not be made complete within
Figure 4. (a) Schematic of a one-bit petahertz optical memory consisting of two basic petahertz devices (LEM-LHM and LHM-LEM devices) and a parallel capacitor with its capacitance \( C \). (b) Write pulse (i.e., an in-plane pulse) to LEM-LHM device casts the charge state in a capacitor from ‘0’ to ‘1’. (c) Read pulse (i.e., an out-of-plane pulse) to LHM-LEM device induces a small charge flow from the capacitor, which reads the original charge state corresponding to \( Q(0) = 0.95 \) or 0.05 to be ‘1’ or ‘0’. (d) Erase pulse (i.e., a strong out-of-plane pulse) to LHM-LEM device exhausts the stored charge in the capacitor, letting the charge state be from ‘1’ to ‘0’. \( E_{\text{bias}} = 0.93 \text{ V} \) (write), \( R_{\text{min}} = 1 \times 10^{-3} \Omega \) (read of ‘1’), \( R_{\text{min}} = 1.9 \times 10^{-2} \Omega \) (read of ‘0’), and \( R_{\text{min}} = 0.5 \times 10^{-4} \Omega \) (erase) are adopted according to the optical-field strengths \( E_{\|} = 2.08 \text{ V} \text{ Å}^{-1} \) (write), \( E_{\perp} = 0.93 \text{ V} \text{ Å}^{-1} \) (read), and 4.17 V Å\(^{-1}\) (erase). \( C \) is taken to be 10 pF.

Figure 5. Write and read processes at \( R_{\text{wire}} \ll R_{\text{min}} \) and \( R_{\text{wire}} \gg R_{\text{min}} \). (a) Write and (b) read processes at \( R_{\text{wire}} = 0 \), and 0.1 \( \Omega \) for \( R_{\text{min}} = 2 \times 10^{-9} \Omega \) (write) and \( R_{\text{min}} = 1 \times 10^{-5} \Omega \) (read) corresponding to \( E_{\|} = 2.08 \text{ V} \text{ Å}^{-1} \) (write) and \( E_{\perp} = 0.93 \text{ V} \text{ Å}^{-1} \) (read). the pulse duration. However, even a wire of a good metal may fail to satisfy \( R_{\text{wire}} \ll R_{\text{min}} \). Hence, a superconducting wire ensuring \( R_{\text{wire}} = 0 \) could be considered. Furthermore, the value of the capacitance \( C \) is assumed to be 10 pF. \( E_{\text{bias}} a (\tau) / 2 = 0.25 \) of figure 4 enables to estimate an approximate amount of stored charges in the capacitor. Knowing \( E_{\text{bias}} \sim 6.5 \times 10^{-5} \text{ V} \text{ Å}^{-1} \) from a \( \sim 5 \text{ Å} \) and \( \tau \sim 1 \text{ ps} \), the voltage \( V \) across

\(^2\) For CaF\(_2\) [29], the minimum value of the transient resistivity \( \rho_{\text{metal}} \) at the metalization under the optical pumping of 2.3 V Å\(^{-1}\) has been reported to be \( \sim 10^{-1} \Omega \) m. Similarly, in the present case, we assume \( \rho_{\text{metal}} \sim 10^{-2} \Omega \) m in a device and may tune \( R_{\text{min}} \sim 10^{-2} \sim 10^{-4} \Omega \) by adjusting the distance between the metal nodes and the field intensity of the pumping pulse. On the other hand, for a wire, a good metal with \( \rho_{\text{wire}} \sim 10^{-8} \Omega \) m could be considered but \( R_{\text{wire}} \) would be \( \sim 10 \Omega \) from \( \rho_{\text{metal}} L_{\text{wire}} / A_{\text{wire}} \) typically with \( L_{\text{wire}} \sim 100 \text{ nm} \) (length) and \( A_{\text{wire}} \sim 10 \text{ nm} \times 10 \text{ nm} \) (cross section). This may fail to satisfy \( R_{\text{wire}} \ll R_{\text{min}} \).
the device would be \( V \sim E_{\text{bias}} \times L_{\text{device}} \sim 0.65 \, \text{V} \) with \( L_{\text{device}} \sim 1 \, \mu\text{m} \). This results in \( Q_C \sim CV \sim 6.5 \times 10^{-13} \, \text{C} \), i.e., corresponding to \( 10^7 \) electrons or holes.

In order for the proposed memory to be realistically meaningful, it should be scalable \([30–32]\).

In figure 6(a), we indicate the feasibility of its integration by laying out four one-bit memory elements in a \( 2 \times 2 \) array. Employing five optical channels, i.e., bit(1), bit(2), write(1), write(2), and read, we attempt a schematic demonstration of four-bit data based on the \( 2 \times 2 \) array. A synchronous application of bit(1) and write(1) pulses given by \( A_1(\tau) \) and \( A_{\perp}(\tau) \), respectively, write ‘1’ in the bit A, whereas a read pulse by \( A_{\perp}(\tau) \) reads out the bits. A schematic example of the four-bit RAM operation under a controlled switch-on of optical channels is demonstrated over three steps in figure 4(b), where the four-bit data are processed as (step I) write ‘1’ in the bit A \( \rightarrow \) write ‘1’ in the bit D \( \rightarrow \) (step II) read out the bits of ‘1001’ \( \rightarrow \) erase the bits \( \rightarrow \) (step III) write ‘1’ in the bit B \( \rightarrow \) read out the bits of ‘0100’. Definitely, the \( 2 \times 2 \) integrated petahertz memory could be generalized to a \( n \times n \) integration, which handles \( n^2 \)-bit data by employing \( 2n + 1 \) optical channels of bit(1), ..., bit(n), write(1), ..., write(n), and read.

3. Conclusions

Hiring a theoretical model of the basic petahertz device made of LHM and LEM dielectrics, we have proposed a superb manipulation of the optical-field-induced petahertz current, which contains a variety of functionalities, e.g., rectifying, switching, amplifying, and suppressing the current. Combining two basic petahertz devices and a parallel capacitor, we have further proposed a one-bit RAM element. Through an investigation of the ultrafast dynamics of fundamental memory tasks such as write, read, and erase processes, we have confirmed that the proposed memory element in fact works at the petahertz clock speed. Finally, we have suggested its scalability or integrability by offering a \( 2 \times 2 \) elemental array consisting of four one-bit memory elements. We believe that the scalable petahertz optical memory, the proposition made here, would be a key ingredient to the new horizon of the future solid-state light-wave electronics.

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