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Optomechanical control of stacking patterns of h-BN bilayer

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Abstract

Few-layer two-dimensional (2D) materials usually have different (meta)-stable stacking modes, which have distinct electronic and optical properties. Inspired by optical tweezers, we show that a laser with selected frequency can modify the generalized stacking-fault energy landscape of bilayer hexagonal boron nitride (BBN). Consequently, BBN can be reversibly and barrier-freely switched between its stacking modes and patterns in a controllable way. We simulate the dynamics of the stacking mode transition with a theoretical model and demonstrate that it happens at picosecond timescale. When BBN is pre-buckled, it can be locked in its metastable stacking modes for a long time. Such a fast, reversible and non-volatile phase transition makes BBN a potential media for data storage.

Electronic and optical properties of few-layer two-dimensional (2D) materials (e.g., graphene1–5, hexagonal boron nitride (h-BN)6–8, transition metal dichalcogenides9–11 and their heterostructures12–15) are strongly affected by their stacking patterns. Some exotic properties are present only for certain stacking sequence16,17. For instance, trilayer graphene in ABA (Bernal) stacking is semi-metallic irrespective of external field; while the semimetallic ABC (rhombohedral)
stacking opens a sizable band gap upon applying a gate voltage\textsuperscript{1–5}. It is thus highly desirable to tune the stacking modes in a controllable way.

Here, we focus on a mechanically and chemically robust 2D material, h-BN, which is an insulator with a wide band gap. Geometrically speaking, the bilayer hexagonal boron nitride (BBN) has five different high symmetry stacking configurations. Among them, $AA'$ (eclipsed with B over N and N over B), $A'B$ (staggered with N over N) and $AB'$ (staggered with B over B) differ only by a translational sliding $u$ along the armchair direction (Figure 1). By using first-principles density functional theory (DFT) calculations (see Methods), we theoretically and computationally suggest that the relative stability of these stacking patterns can be effectively tuned by applying a linearly polarized laser (LPL) with selected frequency. Stacking pattern change corresponds to a slippage of the top layer h-BN with respect to the bottom layer. Since two layers are coupled by weak van der Waals (vdW) interactions, and they have small total mass and are much less constrained by elasticity (compared to martensitic transformations of 3D materials), this stacking glide requires small switching energy and can occur very fast (within picoseconds). The structural change does not accompany with any primary bond breaking or reforming, guaranteeing the reversibility. If BBN is pre-buckled to remove tensile strain and in-plane elasticity, it can be locked in its metastable stacking modes for a long time, making the switch non-volatile.

We use a parameter $u$ (mod 1) to denote these patterns. The $AA'$ configuration is set as $u = 0$, and the $A'B$ and $AB'$ configurations are $u = 1/3$ and $u = 2/3$, respectively. Note that the other two high symmetry stacking modes, $AA$ (eclipsed with B over B and N over N) and $AB$ (staggered) need a $\pi/3$ rotation along the $c$ axis (high energy barrier between them), and will be discussed elsewhere.

We begin our discussions with the generalized stacking fault (GSF\textsuperscript{18,19}) energy landscape along the sliding path calculated by DFT\textsuperscript{20,21} including semi-empirical long range interaction corrections (see Methods). As shown in Figure 2a, $AA'$ is the ground state with lowest energy. The metastable $AB'$ has an energy of 2.1 meV per unit cell (each unit cell contains 4 atoms) higher than that of $AA'$. Between $AA'$ and $AB'$, there are two barriers on two opposite directions (positive and negative $u$). On the positive $u$-direction, the $A'B$ stacking serves as energetic saddle point with 17 meV per unit cell higher than that of $AA'$. The negative $u$-direction barrier at $u \approx 0.8$ (denoted as $SD$ in the following) is 3.0 meV (0.9 meV) per unit cell higher in energy than $AA' (AB')$. These results are
consistent with experimental observations that $AA'$ stacking is more often seen than the $AB'$ stacking on clean samples\textsuperscript{22,23}.

With optical tweezing, one wonders if it is possible to tune the relative stability of these high symmetry stacking modes, and switch them in a reversible and reduced energy barrier (even barrier-free) way. In the following, we will show that it can be done when one imposes a linearly polarized laser onto the sample, with the electric field in the plane of the BN layers. In this paper we do not treat the case of out-of-plane electric fields. Under an electric field $E$ (alternating), the thermodynamic grand potential per unit cell can be written as\textsuperscript{24}

$$G(u | E, \omega) = G(u | E = 0) - E \cdot P_0(u) - \frac{V \varepsilon_0}{2} E \cdot \epsilon^{(1)}(u, \omega) \cdot E$$ \hspace{1cm} (1)

$P_0(u)$ is the intrinsic static polarization, and $\epsilon^{(1)}(u, \omega)$ is the real part of dielectric function of a BBN unit cell with displacement $u$. $V$ is the volume of the unit cell. At zero temperature, $G(u | E = 0)$ is equivalent to the stacking energy per unit cell as shown in Figure 2a, except for a $PV$ term, which is nearly constant for all stacking modes. The second term, which is the first order response to electric field, averages to be zero when the oscillation frequency $\omega$ of the electric field is much greater than the phonon vibrational frequency (usually a few THz). The third term is the second order response to the electric field. The dielectric function $\epsilon(u, \omega)$ should combine the contributions from both ion and electron subsystem. However, the ion subsystem is too slow to follow an ultraviolet laser field, so only the contribution from electron subsystem ($\epsilon(u, \omega) = \epsilon_{\text{electron}}(u, \omega)$) is considered in this work. One notable feature of $\epsilon(u, \omega)$ is that it depends on both the displacement $u$ and the laser frequency $\omega$: the structure determines the dielectric response. Therefore, by careful selection of $\omega$, it is possible to modify GSF energy landscape $G(u | E, \omega)$, and effectively tune the relative stability of different stacking configurations.

We calculate $\epsilon^{(1)}(u, \omega)$ with DFT (see Methods and Supplementary Material). $\epsilon^{(1)}_{xx}(u, \omega)$ for $\omega$ from 4.0 eV to 6.5 eV is plotted in Figure 2b. We can see that in this frequency range, $\epsilon^{(1)}_{xx}(u, \omega)$ is sensitive to both $u$ and $\omega$. For $\omega < 4.0$ eV, which is well below the band edge, $\epsilon^{(1)}_{xx}(u, \omega)$ is nearly constant for all $u$. Two selected frequencies, $\omega = 4.30$ eV and 4.55 eV are shown in Figure 2c. We can see that the dielectric function at a fixed laser frequency is sensitive to the displacement $u$. For $\omega = 4.30$ eV, $\epsilon^{(1)}_{xx}(AB')$ is 19.9, greater than both $\epsilon^{(1)}_{xx}(AA')$ (16.2) and $\epsilon^{(1)}_{xx}(SD)$ (18.0).
Therefore, when a LPL polarized in the $x$-direction with $\omega = 4.30$ eV is applied, the $G(AB')$ would get relatively lower in the GSF energy. When the laser is strong enough ($E > 1.8$ V/nm), $AB'$ stacking can be the most stable, with $G(AA') < G(AB')$. In this case, BBN originally at $AA'$ stacking may slide into $AB'$ stacking. This effect is shown in Figure 2d. In order to induce a barrier-free transition, the minimum electric field strength is $E = 3.5$ V/nm (corresponds to laser intensity $I_0 = 1.6 \times 10^{12}$ W/cm$^2$).

Since $AA'$ stacking is intrinsically stable, the BBN at $AB'$ stacking has the tendency to return to $AA'$. However, as we will clarify later, a domain can be locked in $AB'$ for a relatively long time before collectively transiting to $AA'$. In order to trigger an instantaneous and fast transition from $AB'$ to $AA'$, a laser with frequency $\omega = 4.55$ eV can be applied. We also plot calculated $\epsilon_{xx}^{(1)}(u, \omega = 4.55$ eV) in Figure 2c. It demonstrates an order of dielectric function $\epsilon_{xx}^{(1)}(SD) > \epsilon_{xx}^{(1)}(AA') > \epsilon_{xx}^{(1)}(AB')$. When the laser with electric field strength $E$ greater than 1.0 V/nm is applied, the desired grand potential order $G(AB') > G(SD) > G(AA')$ can be obtained, enabling a barrier free transition from $AB'$ to $AA'$. These alternating electric field magnitudes are moderate and achievable in current experiments.

Next, we explore the dynamics of such transition using a 1D model. As shown in Figure 2e, the model consists 2 layers of particles, each particle represents a unit cell in one layer, and is indexed by $n$ ranging from 1, 2, ... $N$, where $N$ is the total number of unit cells. For simplicity, the positions of particles on the bottom layer are fixed as reference points. Particles on the top layer are allowed to move and their configuration are described by the displacements $\{u_n, n = 1, 2, ... N\}$ relative to the particles on the bottom layer. Three independent interactions are accounted, namely, $U_{\text{elastic}} + U_{\text{vdW}} + U_E$. The first one is the intra-plane interaction on the top layer, which is approximated by a harmonic spring with elastic constant $k$. The second term is the van der Waals (vdW) interaction $U_{\text{vdW}}$ between the two layers. The third interaction is the interaction under the laser field $U_E$, which is determined by the BBN slippage-dependent dielectric function (see Eq. 1).

Now we can put all ingredients together and write down the Lagrangian $\mathcal{L}$ of the system
\[ \mathcal{L} = \int dt \sum_{n=1}^{N} \left[ \frac{ma^2}{2} \left( \frac{\partial u_n}{\partial t} \right)^2 - \frac{ka^2}{2} \left[ (u_{n+1} - u_n)^2 + (u_n - u_{n-1})^2 \right] - U_{\text{vdW}}(u_n) + \frac{\epsilon(u_n)\epsilon_0 V}{2} E^2(n) \right] \]

By applying Lagrange’s equation, we can derive the equation of motion (EOM) for the \( n \)-th particle on the top layer

\[ \frac{\partial^2 u_n}{\partial t^2} = \frac{k}{m} (u_{n+1} + u_{n-1} - 2u_n) - \frac{1}{ma^2} \left[ \frac{\partial U_{\text{vdW}}(u_n)}{\partial u_n} - \frac{\epsilon_0 VE^2(n)}{2} \frac{\partial \epsilon(u_n)}{\partial u_n} \right] - \gamma \frac{\partial u_n}{\partial t} \]  

Here, \( m \) is the effective mass of a unit cell \( (m = m_B + m_N) \), \( a \approx 4.3 \, \text{Å} \) is the lattice spacing (along the armchair direction). The elastic constant \( k \) for a flat BBN can be obtained by fitting the strain-energy curve. In order to calculate the derivative of \( U_{\text{vdW}} \) and \( \epsilon \) with respect to \( u \), we fit them with Fourier series (solid curves in Figure 2a and 2c). The last term in Eq. (3) is a damping term, representative of all possible dissipation effects in the system. \( \gamma \) determines how quickly the oscillations around the GSF energy minimum damp out, and in turn determines the transition time. But the main conclusions do not sensitively depend on the specified value of \( \gamma \). In the following, \( \gamma \) is set as \( 5 \, \text{ps}^{-1} \).

We solve EOM Eq. (3) with a total number of unit cells \( N = 20,000 \). The results (Figure 3) show that \( AA' \) and \( AB' \) can be switched in picoseconds under laser with selected frequency. In Figure 3a, the system is originally in a homogeneous \( AA' \) stacking mode (blue curve). By shining laser with frequency \( \omega = 4.30 \, \text{eV} \), and Gaussian profile \( E(n) = E_0 \exp \left[ -\frac{(n-n_0)^2}{2\sigma^2} \right] \) with \( E_0 = 5.0 \, \text{V/nm} \), \( n_0 = 10000 \) and \( \sigma = 2000 \) (with full width at half maximum around 1 \( \mu \)m. denoted as Laser-A) in the middle region, \( AB' \) stacking domain start to develop. At \( t = 2.5 \, \text{ps} \), Laser-A is turned off and the system is allowed to relax freely. We can see that the middle region of the system first oscillates around \( AB' \) stacking, and after the oscillation damps out, it is locked in \( AB' \) (yellow curve). The whole process takes about 5 ps. The thickness of the domain wall between \( AA' \) and \( AB' \) is about 100 unit cell length, roughly 40 nm. In Figure 3b, the system is initially set as the final state of Figure 3a. At \( t = 0 \), a laser with frequency \( \omega = 4.55 \, \text{eV} \), and the same profile as
Laser-A except that $E_0 = 1.5 \text{ V/nm}$ (denoted as Laser-B) starts to shine on the $AB'$ domain. At $t = 0.5$ ps, Laser-B is turned off, and the system automatically collapsed back to $AA'$. The DWs are erased as well and the system is restored to the initial state in Figure 3a, leaving no trace. That is, by alternatively applying Laser-A and Laser-B, the $AB'$ stacking domain can be written and erased within picoseconds.

In Figure 3c and 3d, we show that the DW can be move towards the $AB'$ ($AA'$) domain by moving Laser-A (Laser-B), with a mechanism similar to writing (erasing) $AB'$ stacking modes. Originally, the system contains a $AB'$ domain, separated from two $AA'$ domains by twin domain walls. In Figure 3c, by shining Laser-A on the left DW and move the laser to left, $AB'$ domain gradually grows to the left, replacing $AA'$ domain. Similarly, in Figure 3d, by shining Laser-B on the right DW and move the laser to left, $AA'$ domain grows to the left and swallows $AB'$ domain. Both processes happen in picoseconds.

It should be noted that in our model system with finite elastic constant $k \sim 10 \text{ eV/Å}^2$, $AB'$ domain is not very stable when the laser is turned off. Due to the elastic interaction between neighboring particles and the intrinsic metastability of $AB'$ stacking mode, $AB'$-$AA'$ domain wall would move towards the $AB'$ direction so that $AB'$ domain shrinks with time, on the order of $\text{nm} \cdot \text{ps}^{-1}$. Therefore, a $AB'$ domain with micrometer dimension would disappear in nanoseconds. However, for a slightly pre-buckled BBN, there is a little bit of out-of-plane displacement and excess area. The excess area can compensate for the in-plane strain induced by stacking incommensurability, possibly at the cost of small out-of-plane displacement. As a result, after the atomic oscillation damps out, all unit cells should be effectively in either $AB'$ or $AA'$ stacking modes with zero in-plane strain, and $AB'$ domain can be locked for a long time. Another way to understand this point is that, the effective elastic constant $k$ for a domain to interact with another domain can be very close to zero, although a single atomic bond still has a non-zero elastic constant. In fact, if we directly set $k = 0$ in Eq. (3) the domain wall has a $\delta$-function shape , and the $AB'$ domain can be locked for infinitely long time provided with zero temperature and no other perturbations (dynamics of the system with $k = 0$ can be seen in Supplementary Materials).

Since the switch between different stacking modes is fast, reversible and non-volatile, we suggest that this BBN could serve as a data storage media. Compared with currently used phase change materials\textsuperscript{25} such as Ge-Sb-Te alloys, which can be switched between crystalline and amorphous
phases upon heating over 1 ~ 1,000 nanoseconds time scale, the non-volatile phase transition in BBN can be ultrafast, athermal, and displacive. The equation-of-motion (3) has similar structure as the Frenkel–Kontorova model, with external forcing by the optical tweezing, that takes advantage of the significant contrast in dielectric response offered by the slippage. The domain walls are topological solitons or dislocations\textsuperscript{26,27}, which are demonstrated to be nucleated and controlled here by light.

Based on this discussion, we illustrate an BBN optical disc drive. BBN stripes are grown on a membrane (Fig. 4), whose two edges are clamped. The membrane has a little slack along the BBN stripe direction, so that BBN stripes are pre-buckled to remove effective in-plane elasticity. Lasers can move around the membrane to read and write. We propose to use high intensity lasers to write and erase AA’ (or AB’) configuration, which can serve as 0 (or 1) state. For reading, one can apply a laser with low intensity, and measure the luminescence, or refractive index by ellipsometry, which is distinct for different stacking modes\textsuperscript{23}. Such a BBN disk can be switched back and forth at THz, much faster than the bit-rates of current phase-change storage media.

We would like to make some final remarks. First, such a scheme of optical tweezing is not limited to 1D slippage: the other two high symmetry stacking modes, AA and AB, can come into play as well. Some experimental and computational works have shown that AB stacking is also stable, with stacking energy comparable to that of AA\textsuperscript{7,22}. The relative stability between these high symmetry stacking modes may be tuned by laser frequency and polarization. This model can thus be easily extended to 2D slippage. With this in mind, one can expect a multiple-bit data storage (“0”, “1”, “2”, and “3”). Second, the spatial density of storage can be very high, with locked-in domain as small as tens of nanometers, and by rolling up the 2D material as a scroll akin to DNA packing, one may achieve high total storage volume in 3D. Third, besides data storage, optical stacking mode transition may find other applications such as in programmable optical switches, phase masks, grating array, etc.

**Methods** Our first-principles calculations are based on density functional theory (DFT)\textsuperscript{20,21} as implemented in Vienna \textit{ab initio} simulation package (VASP)\textsuperscript{28,29}. Exchange-correlation interactions are treated by generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof
(PBE)\textsuperscript{30} form. Core and valence electrons are treated by projector augmented wave (PAW) method\textsuperscript{31} and a plane wave basis set, respectively. The kinetic energy cutoff is set as 400 eV and the first Brillouin zone is sample by a11 × 11 × 1 Γ-centered \textbf{k}-point mesh. Interlayer van der Waals interactions are treated with DFT-D3 method of Grimme with Becke-Jonson damping\textsuperscript{32,33}. For each displacement \( u \), the in-plane lattice constant is fixed at 2.50 Å but the inter-plane distance is allowed to relax, until forces on all atoms are smaller than 0.001 eV/Å. The dielectric function is calculated in the independent particle approximation (IPA). More accurate quasi-particle GW\textsuperscript{34,35} with exciton binding correction (Bethe-Salpeter equation, BSE\textsuperscript{36,37}) calculations are also performed and the results are qualitatively consistent with IPA results.

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![Figure 1](image)

Figure 1: Three high symmetry stacking modes $AA'$, $A'B$, and $AB'$ of BBN. They are different by a translational sliding $u$ along the armchair direction (red arrow). $AA'$, $A'B$, and $AB'$ correspond to $u = 0, 1/3$ and $2/3$, respectively.
Figure 2 (a) Intrinsic BBN GSF energy landscape along the armchair direction. Three high symmetry stacking modes are labelled. $AA'$ is stable, $AB'$ is metastable, and $A'B$ is unstable. $SD$ denotes the saddle point from $AA'$ to $AB'$ in the negative-$u$ direction. (b) Calculated dielectric function $\epsilon_{xx}^{(1)}(u, \omega)$ for $\omega$ from 4.0 eV to 6.5 eV. (c) Dielectric function $\epsilon_{xx}^{(1)}(u, \omega)$ at two selected frequency $\omega = 4.30$ eV and 4.55 eV. $\epsilon_{xx}^{(1)}(u)$ at a fixed frequency is dependent on the displacement $u$ and can tune the shape of stacking energy landscape $G(u)$. $\omega = 4.30$ eV favors $AB'$ stacking while $\omega = 4.55$ eV favors $AA'$ stacking. In (a) and (b), crosses are from DFT calculations while solid curves are Fourier series fittings. (d) GSF energy $G(u, \omega = 4.30$ eV) at several different laser intensity. $I_0$ corresponds to $E_0 = 3.5$ V/nm and is strong enough to induce barrier free transition from $AA'$ to $AB'$. (e) the 1D model system as described in the main text. Each pink point represents a unit cell on one layer.
Figure 3 Dynamics of the model system under different laser illumination. (a) **Writing $AB'$ domain.** Initially the system is in a homogeneous $AA'$ stacking mode. From $t = 0$ to $2.5 \, \text{ps}$, Laser-A, which favors $AB'$ stacking mode shines on the middle region. An $AB'$ domain gradually develops. After several oscillations, middle region of the system is locked in the $AB'$ stacking mode. (b) **Erasing $AB'$ domain.** The initial state here is the final state in (a). Between $t = 0$ to $0.5 \, \text{ps}$, Laser-B, which favors $AA'$ stacking shines on the $AB'$ domain. The system is restored to the initial state in (a), which is a homogeneous $AA'$ domain. (c, d) **Moving DW.** Laser-A (Laser-B) shines on the DW between $AA'$ and $AB'$ domain, and moves towards $AB'$ ($AA'$) domain. The DW moves in the same direction as the laser.
Figure 4 A schematic sketch of the BBN disk. BBN stripes are grown on a membrane. The membrane is clamped on two edges and has a little slack, so BBN stripes are slightly buckled. Laser moves around to read/write data on the BBN stripes. Digital 0/1 can be represented by $AA \perp AB'$ stacking modes.
Supplementary Materials
Figure S1 Dynamics of the model system under laser illumination. The laser has the same time and spatial profile as that in Figure 3. But the force constant $k$ in Eq. (3) is set as zero, thus the DW is thinner and should have $\delta$ function (zero width) form as time approaches infinite. The DW, once formed, has no translational movement with time.
Figure S2 Dielectric function $\varepsilon_{xx}^{(1)}(u, \omega)$ from GW-BSE calculation. Two selected frequency $\omega = 4.95 \text{ eV}$ and $5.15 \text{ eV}$ favor $AB'$ and $AA'$ stacking, respectively.