Light-responsive organic flashing electron ratchet

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Contributed by Mark A. Ratner, July 5, 2017 (sent for review April 11, 2017; reviewed by Ignacio Franco and Eran Rabani)

Ratchets are nonequilibrium devices that produce directional motion of particles from nondirectional forces without using a bias, and are responsible for many types of biological transport, which occur with high yield despite strongly damped and noisy environments. Ratchets operate by breaking time-reversal and spatial symmetries in the direction of transport through application of a time-dependent potential with repeating, asymmetric features. This work demonstrates the ratcheting of electrons within a highly scattering organic bulk-heterojunction layer, and within a device architecture that enables the application of arbitrarily shaped oscillating electric potentials. Light is used to modulate the carrier density, which modifies the current with a nonmonotonic response predicted by theory. This system is driven with a single unbiased sine wave source, enabling the future use of natural oscillation sources such as electromagnetic radiation.

Biological environments are noisy, chaotic, and highly damped (1). Transporting particles under those circumstances is a challenge, for which nature developed molecular motors, such as the myosin–actin system responsible for muscle contraction (2), the kinesin molecular walker (3), and ATP synthase (4). Such systems couple inherent structural asymmetries and relaxation with nondirectional sources of energy, like chemical energy, to obtain directional motion in the presence of strong damping and thermal noise through a mechanism called “ratcheting” (5, 6). For example, in the myosin–actin system, thermal fluctuations of the myosin head on an elastic tether lead to occasional binding of the head to an actin filament, at which point thermal energy is transduced to elastic energy, and the filaments translate relative to one another. A chemical reaction-coupled conformational change in the myosin head upon translation induces release of the head from the actin filament, and renders the translation irreversible. The system thereby uses a chemical trigger to rectify random thermal motion (7). The design principles of natural systems are today being used to develop a variety of molecular machines (8) and to achieve, experimentally, ratcheting of micrometer-sized particles (9), DNA (10), and cold atoms (11).

The concept of an electron ratchet has been explored theoretically (12–14) and, in rare cases, experimentally (15–21). There are two major types of electron ratchets: “flushing,” in which the electron moves along a periodic potential surface with locally asymmetric repeat units that oscillates between two states, while the source-drain bias along the direction of transport is constantly zero (5, 9); and “tilting,” in which the shape of the potential surface remains constant, but the source-drain bias oscillates with a time average of zero (5). Fig. 1 shows a mechanism of transport in perhaps the simplest 1D flashing ratchet system, an “on/off” ratchet. Our interest lies in adapting the flushing ratchet mechanism to transport electrons within highly scattering, low-conductivity environments, such as an amorphous organic or nanostructured material. We focus on flashing ratchets because they do not require a source-drain bias, but are rather based on local fluctuations of the potential energy surface within the transport material, achievable using stimuli such as heat and light (our prototype system uses an AC electric field), and, in this way, are more analogous to biological ratchets. Ratcheting could not only provide unexplored modes of transport in materials that are insulating relative to crystalline semiconductors but also result in unique current–voltage characteristics and responses to external stimuli for both electronic and energy conversion applications.

In 2005, Müller et al. (18) used asymmetrically spaced pairs of electrodes, which create a simple sawtooth-shaped potential, under a cryogenically cooled 2D electron gas (2DEG) layer, to produce the first flashing electron ratchet. In 2011, Roeling et al. (19) demonstrated the first room temperature flashing electron ratchet, using electrode pairs under a layer of pentacene, an organic semiconductor. In 2013, Tanaka et al. (20) replaced the electrode pairs with flat, asymmetric electrodes, to ratchet electrons in a 2DEG nanowire. These demonstrations of electron ratcheting in conducting and semiconducting materials are remarkable, and inspire many still open questions regarding the mechanisms by which the time-dependent potential surface introduces the asymmetry necessary to create directional transport; these mechanisms are complex and often unintuitive (5), especially with regard to low-mobility, highly damped conditions.

Here, we demonstrate room temperature electron ratcheting in a blend of poly(3-hexyl-thiophene-2,5-diyl) (P3HT) and [6,6]-phenyl-C61 butyric acid methyl ester (PCBM) (μ ~ 10−3 to 10−4 cm2·V−1·s−1), and identify some fundamental behaviors of an organic electron ratchet that are critical for its useful application. The major advances of this work are as follows: (i) Previous demonstrations required the time oscillation of the potential to follow a biased waveform [e.g., on/off or sin(ωt)] in our design, even an unbiased waveform can produce transport, because the effective potential is biased in the z direction (through the thickness of the transport layer, Fig. 2A), due to the dielectric response of the layer, which causes the applied field to decay with distance from the electrodes. This result introduces the possibility of using natural oscillation sources (e.g., electromagnetic radiation), which are typically unbiased sine waves. (ii) In previous work, only a single, asymmetric electric field was used; here, we demonstrate ratcheting with a symmetric field, which is a significant result because it demonstrates ratcheting with a featureless bias field, and eliminates the need for a field-induced bias. (iii) We demonstrate operation of the ratchet over a significant range of optical intensities, extending the range of applications of this system.

Significance

Transport of objects in man-made systems typically relies on energy gradients that span the distance over which the particles must travel. Biological systems do not have these large-scale gradients available and instead transport particles with repeated local interactions between asymmetric structures, powered by nondirectional sources of energy like heat or chemical reactions. This process is called “ratcheting.” Here, we exploit the ratcheting mechanism to transport charge-carrying particles, electrons, through an amorphous organic film in which local structural inhomogeneities disrupt the gradients needed for typical electrical conduction to occur efficiently. This work explores basic mechanisms by which energy that is normally wasted in an electronic or photoelectrical device could be harnessed to do electrical work.

Author contributions: O.K., B.L., M.A.R., and E.A.W. designed research; O.K. performed research; O.K., B.L., M.A.R., and E.A.W. analyzed data; and O.K., B.L., M.A.R., and E.A.W. wrote the paper.

Reviewers: I.F., University of Rochester; and E.R., University of California, Berkeley.

The authors declare no conflict of interest.

Data deposition: The measurement data supporting this work are available from the Northwestern University Libraries Archive (https://doi.org/10.21985/N0KD32).

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This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1705973114/-/DCSupplemental.
simple sawtooth potential shape was applied, but our asymmetrically shaped electrodes allow us to access a variety of shapes. We use this capability here to identify general, shape-independent behaviors of the ratchet device, and it could be used in future work to explore the parameter space of potential shapes. (iii) Implementing ratchet mechanisms in useful devices would ideally mean using high particle (e.g., electron) concentrations. We use a photoresponsive material to modulate the carrier density in situ and find a non-linear, nonmonotonic dependence of the ratcheting current on the charge carrier density, consistent with theoretical predictions (22–24) made for Brownian particles. In many cases, we find that increased carrier density actually reduces the ratchet current; this result suggests a significant role for interparticle interactions in ratchet operation.

Although our flashing ratchet system is used here as an investigative tool, it is an active device—we show that ratcheted electrons do work against a bias and provide a measurement of power conversion efficiency for an electron ratchet.

**Experimental Design**

The ratchet is configured similarly to a field-effect transistor in the coplanar (bottom contact) bottom-gate configuration (Fig. 2 A–C and SI Appendix, Figs. S1 and S2), with a channel length of 12 μm and width of 500 μm (note that the views in Fig. 2 A and C are rotated 90° in the x–y plane from the view in Fig. 2B). The transport layer is a 200- to 400-nm-thick, thermally annealed bulk heterojunction (BHJ) film [1:1 (wt/wt) blend of P3HT: PCBM], where electron transport is mediated primarily by PCBM and hole transport primarily by P3HT (25). We use this photoactive blend as it allows us to modulate the charge carrier density in situ using illumination and provides a particularly damped environment for the electron (an undoped, primarily amorphous organic film with high interfacial area between P3HT and PCBM) (26). Although both electrons and holes are present in the system, given the large Schottky barriers that form between PCBM and our Ag source and drain electrodes (27), the current we extract comes almost exclusively from mobile holes. There is no applied source-drain bias or equivalent difference in the work functions of the identical source and drain electrodes, and no carrier-blocking interfacial layers or chemical dopants that introduce permanent gradients (Fig. 2A); this device is not a solar cell, nor even a diode. Rather, it is designed such that directional current is only achieved by ratcheting electrons along an oscillating potential surface created by an array of eight “finger” electrodes (FEs) with an asymmetric thickness profile in the direction of transport, fabricated using focused-ion-beam–assisted deposition, Fig. 2D. The FE shape, together with the dielectric response of the transport layer, determine the shape of the electric field (Fig. 2 E and F), denoted \( G(x,z) \). By oscillating the potential applied to the FEs as \( F(t) = V_0 + A \sin(2\pi ft) \), where \( A \) is the amplitude, \( f \) is the flashing frequency, and \( V_0 \) is a constant offset, we obtain a multiplicative effect.
flashing ratchet potential, \( V(x, z, t) = G(x, z)F(t) \). In all results shown below, \( V_0 = 0 \) V, unless otherwise noted, and thus the mean applied potential in all parts of the film is zero.

**Results**

We fabricated 25 devices, with various FE shapes (SI Appendix, Fig. S4), and all were active ratchets, producing frequency-dependent source-drain currents on the order of 1–10 nA at zero bias across the device (in the dark or under illumination), but only in the presence of an oscillating potential. Fig. 3 A–C shows the FE profiles [using atomic force microscopy (AFM)] and the dependence of the short-circuit current, \( I_{sc} \), on the frequency of oscillation for a few representative devices; we discuss this dependence below. These \( I_{sc} \) values are very close in magnitude to those measured in P3HT:PCBM devices in a similar FET configuration under a \( \sim 50-100 \text{-} \text{mV} \) source-drain bias (with zero gate potential) (27). The responses we observe are consistent with the known and theoretically predicted characteristics of flashing ratchets, and inconsistent with alternative mechanisms, as we discuss in SI Appendix, section 2. Our tunable electrode shape allows us to apply a wide range of ratchet potential shapes, and thus document general behaviors, which will facilitate future work on the complex problem of linking ratchet performance to the shape of the potential (14, 28).

**Dependence of Ratchet Current on the Oscillating Frequency of the Applied Potential.** In a flashing ratchet, the oscillating potential not only maintains the system far from equilibrium but also alternately decelerates and accelerates the charge carriers so that they can relax asymmetrically along gradients in the applied potential and produce current (14, 29). The frequency of switching must be comparable with some characteristic timescale(s) of electron motion (30), or else the electron will remain permanently trapped (low frequency) or see a static average potential (high frequency), both of which result in zero current. In devices in our devices, regardless of potential shape, the measured current strongly depends on the frequency of the flashing potential, and we observe the strongest currents in the 100-kHz to 1-MHz range (Fig. 3 B and C, and SI Appendix, Fig. S13), which is reasonable given the travel time between potential wells for carriers under the applied field calculated from the known carrier mobilities in P3HT:PCBM (SI Appendix, section 3.3). In some cases, the short-circuit current reverses direction with changes in amplitude or frequency, a remarkable characteristic of ratchets (5) (Fig. 3C, traces 4 and 6, and SI Appendix, Fig. S15) that depends on the shape of the potential surface and differentiates them from pumps and diode rectifiers.

Devices incorporating P3HT:PCBM blends ranging from 50% to 100% P3HT all exhibit ratcheting. Although we have not explored the composition dependence of the ratchet performance in detail, we did observe that (i) the DC conductivity of the films decreases with increasing P3HT content, and (ii) with decreasing conductivity, both the ratchet (short-circuit) current and the oscillation frequency that produces the peak ratchet current (SI Appendix, Fig. S9) decrease, consistent with a dependence of this peak frequency on the timescale of electron motion in the material.

**Ratcheting with Unbiased Oscillation of the Potential.** An unbiased waveform, such as a sine wave, includes equal durations of equal magnitude positive and negative states, and so its time-averaged value is zero. A ratchet that operates using an unbiased waveform as an energy input is more widely applicable than one that uses a more specialized waveform, as sine waves are the simplest waveform to produce electronically and can be harvested from electromagnetic radiation. To use incoherent natural radiation sources, a design closer to a temperature ratchet might be more appropriate (31). Our devices produce net transport for unbiased waveforms, including triangle, sine, and square waves (Fig. 3B). This result is not trivial; to understand why, let us describe the forces acting on the carriers using the Hamiltonian \( H = H_0 + G(x)F(t) \). To produce a current, the Hamiltonian must contain an asymmetry in the \( x \) direction. The system Hamiltonian \( H_0 \) accounts for random static

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Fig. 3. Influence of the temporal waveform on ratchet current. (A) AFM line scans of four devices. (B) Bidirectional sweeps of \( I_{sc} \) vs. flashing frequency in the dark for three unbiased temporal waveforms (indicated), for device 5, \( A = 10 \) V; the triangle waveform is only applied up to 500 kHz due to instrument limitations. The dashed line is the result of an analytical model for a square-waveform drive (see text); (C) \( I_{sc} \) vs. flashing frequency, for devices 1–6 (Figs. 3A and 4A), \( A = 10 \) V; we consistently observe somewhat unstable currents at the low end, and a resonance peak at the high end (shaded area). We do not consider signals in these regions when determining peak ratchet frequency and current. (D) \( I_{sc} \) vs. amplitude for device 1 in Fig. 4A, in the dark, for various frequencies, with the trace for 50 kHz in red.
and dynamic disorder in the system, and is nominally isotropic; the spatial potential $G(x)$ is asymmetric, but when multiplied by a periodic antisymmetric function $F(t)$ (e.g., a sine wave), the carriers will alternately experience either $G(x)$ or $-G(x)$, which possess exactly opposite asymmetries. The time-averaged Hamiltonian is spatially symmetric and can produce no current (29). All previous flashing ratchet studies have used temporally biased waveforms to drive the ratchet potential, such as a squared sine wave (19) or an on/off drive (10, 20); such a biased waveform can be described as $F_1(t) = F_0 + F(t)$, resulting in the Hamiltonian $H = H_0 + G(x)F(t) + G(x)F(t)$, where a time-invariant asymmetric term, $G(x)F(t)$, exists, and a current can be produced. In a typical experimental implementation, pairs of electrodes are driven by two phase-shifted AC sources, each producing a sine(t) signal (10, 18, 19). If a permanent asymmetry is built into the system, for example, using an ion-gradient in the transport layer, a pump can be produced, which will operate using unbiased sine waves (29), but will not display a peaked dependence of the current on frequency, or current reversals (32), some typical features of a flashing ratchet (5).

Our ability to rectify AC fields with unbiased waveforms relies on two facts: (i) the potential, applied by FEs under the transport layer, decays with distance from the FEs, is thus asymmetric in the $x$ direction, and the carriers can move in that direction, due to the nonzero thickness of the transport layer; and (ii) the potential switches between fully positive (for holes, repulsive) and fully negative (attractive) states (Fig. 2F), in contrast with the potentials used in previous studies. The potential can thus oscillate the carriers up and down in the transport layer. The potential both decays and becomes more symmetric with distance from the FEs (Fig. 2F), so the potentials the carriers experience at the bottom and top boundaries of the transport layer are not mirror images, and an asymmetry is maintained, even for an unbiased temporal waveform. In fact, we can adequately simulate the frequency dependence for our ratchet current using an analytical model developed by Rozenbaum (33) (e.g., Fig. 3B, dashed trace), where noninteracting particles traveling in 1D experience a potential oscillating between two states of opposite polarity and unequal magnitudes (resulting, in our system, from the aforementioned decay in magnitude and asymmetry throughout the nonzero thickness of the transport layer). The model is detailed further in SI Appendix, section 3.7. In our experiment, then, movement in the $z$ direction with unbiased driving produces a similar result to a 1D ratchet with biased driving.

The additional asymmetry provided by motion of the electrons in the $z$ direction was not available in previous experimental systems, which used either a 2D electron gas (18, 20), or pentacene (19). The latter required a static gate bias to create a thin conductive channel, which effectively limits conductance to a 2D sheet. These authors therefore did not report transport for unbiased waveforms. Our devices are not limited to unbiased waveforms and, for instance, produce similar currents when we add a constant offset $V_0$ to the temporal sine wave, even to the limits of $V_0 = \pm A$ (oscillating the potential from 0 to 2A or 0 to −2A) (Fig. 3 and SI Appendix, Fig. S6D).

**Dependence of Ratchet Current on the Amplitude of the Applied Potential.** We observe a power law dependence of $I_{sc}$ on the amplitude of the potential, $A$ ($I_{sc} \propto c A^c$, where $c$ is a constant), at low amplitudes (Fig. 3D and SI Appendix, Fig. S14): for all devices, amplitudes of less than ~2 V are too weak to effectively trap and accelerate the charge carriers, and produce no current. Higher amplitudes increase the current in general, but for some devices and frequencies (e.g., 50 kHz in Fig. 3D), the current saturates at high $A$, producing a sigmoidal shape; this observation is consistent with the theoretical prediction that the current is a peaked function of $A$, appearing sigmoidal at low $A$ (34). This sigmoidal dependence stands in contrast with the linear dependence of diode rectifiers (32). The ratcheting process relies on periodically localizing carriers in potential wells, and higher amplitudes can do so more effectively, especially for relatively slow oscillation frequencies, where the trapping period is long. However, for the low carrier concentration in organic semiconductors, one is limited by the number of available carriers, when a stronger field cannot trap any more carriers per well, and cannot therefore increase the current.

**The Dependence of Ratchet Current on Carrier Density.** Using photocitation, we modulate the carrier density in situ, without modifying the device structure, the chemistry of the film, or applying a static gate potential, which would modify the applied ratchet potential. Fig. 4 shows the shapes of the FE arrays for devices 1 and 2, and their photoelectrical characteristics, under short-circuit (zero source-drain bias) conditions. Interestingly, we observe that illumination can either enhance or diminish the
ratchet current relative to its magnitude in the dark (Fig. 4 B and C),
depending on the specific device, even though the light is certainly
creating more charge carriers, as evident from the increased conduc-
tivity (Figs. 4E and 5A, “Control” curves). The magnitude of the
photoresponse of the ratchet current $(I_{sc}(\text{light}) - I_{sc}(\text{dark}))$
depends on the wavelength of illumination, roughly according to the BHJ’s
absorption spectrum (Fig. 4E). The photoresponse saturates around
$\sim 30 \text{ mW/cm}^2$ for both devices (Fig. 4C).

Our findings confirm previous theoretical predictions for a
nonlinear, nonmonotonic dependence of $I_{sc}$ on carrier density
(22–24), based on that fact that, in a ratchet, carriers are peri-
odically localized in potential wells, as opposed to the continuous
flow generated by a bias. As the carrier density increases, (i) $I_{sc}$
will trivially increase due to the higher number of carriers; (ii)
the fields created by the carriers can provide an additional
driving force to enhance transport, and (iii) interparticle re-
pulsion makes trapping during the attractive portion of the trajec-
try more difficult, and impedes transport. The exact value at
which increasing the carrier density decreases the current
depends on the specific shape of the potential wells, and the initial
density of carriers (controlled by the precise structure, doping
and thickness of the transport layer), and so varies between
devices. Our results emphasize the potential importance of car-
rier–carrier interactions in ratchet operation. We believe that the
$I_{sc}$ of devices 1 and 2 respond oppositely to light because the
devices sit at different points on the theoretically predicted (22–
24) peaked function describing current vs. carrier density, or,
alternatively, that the specifics of the peaked function differ
based on the potential shape. The observed photoresponse,
which varies drastically with the frequency and amplitude of the
potential, and the intensity and wavelength of the light, raises the
possibility of using ratchets as photoswitches (Fig. 4D) in
electrooptical circuits. Based on the measured photoresponse, the
operating parameters of a given ratchet can be selected so that
illumination can increase, decrease, or even reverse the direction
of the current (e.g., SI Appendix, Fig. S15), making it a highly ver-
satile component; the highest on/off ratio we have recorded is
$\sim 10$ (SI Appendix, Fig. S15E).

The Electron Ratchets Do Work. Up to this point, we have focused
on measuring the current at zero source-drain bias. Applying a
bias allows us to characterize the conductivity and the open-
circuit voltage, $V_{oc}$, produced by the ratchet. First, without appl-
ying an oscillating potential to the FEs, we observe a linear $I$–$V$
relationship over the small voltage range used in all cases, passing
through the origin (Fig. 5, “Control” curves). Illumination in-
creases the conductivity for all of the devices, but, even under
illumination, none produces open-circuit voltage, $V_{oc}$, or $I_{sc}$,
without an oscillating potential, an expected result given that the
device is not a diode. When a flashing potential is applied, the
conductivity of the devices increases (Fig. 5, “Ratchet” curves,
Fig. 4E, and SI Appendix, Fig. S16), and the devices produce
nonzero $I_{sc}$ and $V_{oc}$ and therefore transport charge carriers
against an applied bias. Illuminating the device always increases
the film’s conductivity, but can either increase or decrease the
magnitudes of $I_{sc}$ and $V_{oc}$ (depending on the specific device, Figs.
4 and 5) produced under ratcheting conditions. $I_{sc}$ and $V_{oc}$ are
always of opposite signs and vary in concert in response to illu-
mination and oscillation frequency (SI Appendix, Fig. S13C).
Even for cases where illumination decreases $I_{sc}$ and $V_{oc}$, there
even exist other values of the source-drain bias where illumination
increases the current, and one can set specific bias values so as to
use light to turn the ratchet current on or off (Fig. 5B, Inset).

Although our ratchets are intended to elucidate mechanisms,
rather than produce power, it is interesting to calculate their internal
energy efficiency. For device 1 (Figs. 4A and 5), the maximum DC
power output ($P_{out} = I \cdot V$) is $\sim 0.64$ nW. To quan-
tify the power input, we treat the system as a capacitor,
formed between the FE array and the transport layer (Fig. 2C).
The applied potential charges and discharges the capacitor, thus
investing and regaining energy, and the loss in that process is

$$P_{loss} = V_{rms}^2 \cdot 2f \cdot C \cdot DF,$$

where $V_{rms}$ is rms voltage, $f$ is oscillation frequency, $C$ is capacitance, and $DF$ is dissipation factor (35). Power is lost to polarize the dielectric and transport layers, and
accelerate carriers in both desired and undesired directions. The
power loss is therefore the baseline amount of power needed to
maintain the oscillating field, disregarding loss in external
circuits like the frequency generator. We measured $C$ and $DF$
for the frequencies used in this study for multiple devices (SI
Appendix, Fig. S5 C and D); typical values are $C = 1.3$ pF and
$DF = 0.25\%$. For illuminated samples, we include the energy
contributed by the absorbed light (which creates photogener-
carriers, but also dissipates unproductively as heat), so that $P_{in} = P_{loss} + P_{light}$. For example, for $A = 10$ V and $f = 500$ kHz,
and $P_{loss} = 510$ nW. The 532-nm laser at 4.5 mW contributes
$\sim 380$ nW (SI Appendix, section 3.4), so the efficiency of device
1 in Fig. 5 is $\eta = P_{out}/P_{in} = 0.07\%$. The highest efficiency we

![Fig. 5. Producing power. (A) $I$–$V$ scans of device 1 with (“32 nm”) and
without (“dark”) illumination, and with (“Ratchet”) and without (“Control”)
the flashing potential; the device only produces an $I_{sc}$ and an open-circuit
voltage $V_{oc}$ with the flashing potential. In the lower-right quadrant
(shaded), the ratchet is doing work; $f = 500$ kHz, $A = 10$ V. (Inset) DC power
($P_{out} = I \cdot V$) produced by the ratchet, with and without illumination.
(B) Current–voltage ($I$–$V$) scans, as in A, for device 2, $f = 200$ kHz, $A = 10$ V.
(Inset) A section of the $I$–$V$ curves highlighting biases where illumination
turns the ratchet current on (1) or off (2).]
measured was 0.49%, for a device in the dark that produces its peak current at a lower frequency, lowering \( P_{\text{loss}} \) (SI Appendix, Fig. S17 C and D). This experimental efficiency value is similar to those estimated by Roelting et al. (19), using a combination of simulation and experiment, for an organic flashing ratchet (0.7–1.5%). The efficiency could be improved by, for example, depositing the BHJ selectively over the channel using inkjet printing, which decreases the dissipation factor and capacitance, and, in our preliminary work, reduces the power loss by a factor of 8 on average. Modifying the temporal oscillation of the potential by, for example, changing the duty ratio of a square wave drive, also increases the output power (SI Appendix, Fig. S17 E and F).

Conclusions

We have constructed room temperature flashing electron ratchets from a blend of conjugated organic materials; these devices produce a source-drain current in the absence of a source-drain bias, and produce power, upon the application of an oscillating potential. Although we focus here on understanding mechanisms of ratchet operation, with minimal optimization of current or efficiency, the ratchets we have built could rectify radiowaves to remotely power nanoelectronic circuits (36, 37) if combined with an antenna. They also produce enough current and voltage to act as passive logic elements within optoelectronic circuits (38), for instance by using one ratchet to light a micro-LED, which in turn controls the output of another ratchet, exploiting the sensitivity of the electrical characteristics of our ratchets to light. Tunable, stimulus-responsive prototype systems such as ours, capable of testing theoretical hypotheses regarding ratchet mechanisms, are necessary to transform electron ratchets from fundamentally interesting implementations of a process nature has optimized to a set of useful technologies.

Methods

Device Fabrication. Devices were fabricated on 2" p-type (n-doped) 300-μm-thick Si wafers with 1,000-nm thermal oxide. Using photolithography, 10-nm Ti/40-nm Au contact lines and pads for the FEs were deposited. Focused-ion beam was used to deposit eight Pt FE tips with an asymmetric thickness profile, 400-nm wide, 572.5-μm long, and ∼50-nm thick, spaced 400 nm apart. A dielectric layer of 13-nm plasma-enhanced chemical vapor deposition (PECVD) SiO\(_2\)/124-nm spin-on-glass (for surface planarization)/20-nm PECVD SiO\(_2\) was then deposited. Using photolithography and reactive ion etching, the FE contact pad was exposed. Photolithography was then used to deposit, on either side of the FE array, source and drain electrodes, 50-nm thick (10-nm Ti/40-nm Ag), 500-μm long, 12-μm apart, with contact lines and pads. The BHJ layer was deposited from a 10 or 5 mg/ml (for the series of different blend ratios) solution of d-ochlorobenzene with 3% 1,8-dioctooctane, using spin coating or drop casting, followed by annealing at 110 °C for 10 min. The deposition and annealing were performed in ambient air.

Characterization. FE arrays were imaged using an atomic force microscope; the dielectric layer was measured using a spectroscopic ellipsometer and a reflectometer. Contacts were made using a Signatone S-1160 probe station, and the current was measured using a Keithley 6430 electrometer, with or without a premultiplier; a Keysight B2902A SMU; or an Agilent 4155C parameter analyzer, all producing the same results. The FE-common bias was modulated using a Tektronix AFG3022C function generator. The capacitance and dissipation factor were measured using an Agilent 4285A LCR meter.

Illumination. Unless otherwise noted, scans were performed in the dark. Where stated, devices were illuminated using 4.5-mW laser diodes at 808, 635, and 532 nm. In some cases, devices were illuminated with a 150-W halogen illuminator with 10 ± 2-nm FWHM bandpass filters and peak transmittance wavelengths of 400 ± 2, 450 ± 2, 500 ± 2, 550 ± 2, 600 ± 2, 650 ± 2, and 700 ± 2 nm. A more detailed methods section is presented in SI Appendix.

ACKNOWLEDGMENTS. We thank Dr. Mario Tagliazucchi, Dr. Igal Szleifer, and Dr. Mark C. Hersam for helpful discussions. This material is based upon work supported as part of the Center for Bio-Inspired Energy Science, an Energy Frontier Research Center funded by the US Department of Energy, Office of Science, Basic Energy Sciences under Award DE-SC0000989. This work used Northwestern University Micro/Nano Fabrication Facility (NUFAB), supported by the State of Illinois, the Materials Processing and Microfabrication Facility at Northwestern University, supported by the Materials Research Science and Engineering Center (MRSEC) program of the National Science Foundation (NSF) (DMR-1121262), and the Electron Probe Instrumentation Center (EPIC), Scanned Probe Imaging and Development (SPID), and Keck-II facilities of the Northwestern University Atomic and Nanoscale Characterization Experimental Center (NUANCE), which receives support from the Soft and Hybrid Nanotechnology Experimental Resource (NSF Grant NNC-1542205); the MRSEC program; the International Institute for Nanotechnology (IIN); the Keck Foundation; and the State of Illinois.