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Electron Ratchets: State of the Field and Future Challenges

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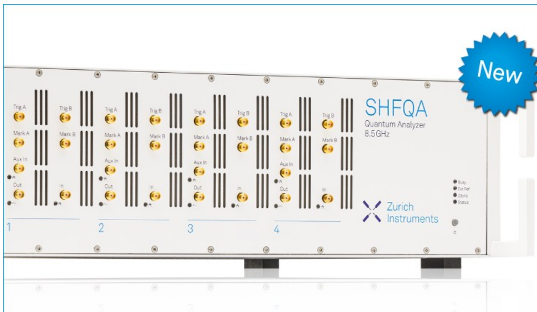

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ABSTRACT

Electron ratchets are non-equilibrium electronic devices that break inversion symmetry to produce currents from non-directional and random perturbations, without an applied net bias. They are characterized by strong parameter dependence, where small changes in operating conditions lead to large changes in the magnitude and even direction of the resulting current. This high sensitivity makes electron ratchets attractive research subjects, but leads to formidable challenges in their deeper study, and particularly to their useful application. This perspective reviews the progress that was made in the field starting from the first experimental electron ratchets in the late 1990s, and how the field spawned multiple designs with very different properties. We discuss the possible uses of electron ratchets in sensing and energy harvesting, and the specific issues encountered when idealized behavior meets complex reality. We promote an application-driven approach where complexity is not necessarily detrimental and argue that a system level perspective would be beneficial over reductionism. We highlight several promising research directions, which revolve around the intentional study of complex effects, and the modeling of realistic devices.

INTRODUCTION

In this perspective, we discuss the origins, development, current state, and research directions of the electron ratchet field. We introduce the concept of an electron ratchet, and ratchets in general, by examining in detail the first electron ratchet papers. From this concept emerge two uses for electron ratchets, sensing and energy harvesting, which depend on controlling current reversals, a defining feature of ratchets. We review experimental and theoretical progress on understanding the general properties of electron ratchets. The sensitivity of the ratchet current to variations in any parameter leads us to advocate for an application driven approach to future research. We conclude by highlighting major challenges and opportunities for the field of electron ratchets. While we strive to present the major advances in the field, this work is not intended to serve as a comprehensive review, so we limit our discussion to the explicit transport of electrons, rather than all ratchets with electronic effects. We, thus, exclude ratchets transporting vortices in superconductors,[1] spin,[2] magnetic flux quanta,[3] or magnetic domain walls.[4]

To properly introduce the ratchet field, we must delve into 20th century thermodynamics, and particularly the apparent paradoxes surrounding the second law of thermodynamics. A major question in 19th and 20th century thermodynamics was whether one can obtain something for nothing—extract energy from seemingly useless fluctuations, such as thermal noise. Maxwell's Demon (or Daemon) is a particularly well-known thought experiment designed to test this question; the paradoxical nature of the demon, where the entropy of the system is seemingly decreased without expending energy to do so, was addressed in multiple ways over the years.[5] Explanations focused on the inclusion of the demon as an integral part of the system, and on the energy cost of measuring the velocity of the particles. More specifically, Rolf Landauer showed that while measurements can be done in a reversible manner (and thus not increase entropy), eventually the demon would need to erase old velocity measurements to make room for new ones, and the erasure is an irreversible process, which increases the overall entropy.[6]

A real, rather than fanciful, mechanism for rectifying motion is the mechanical ratchet, where the rotation of a gear with asymmetric teeth is checked by a pawl, which imposes unidirectional rotation; clocks use ratchets to translate the oscillations of a pendulum into the unidirectional motion of the hands. Maxwell's demon was on physicists' minds, and understanding the second law of thermodynamics was a major issue, as its statistical nature raised serious objections. Around 1900, Gabriel Lippman proposed the use of a ratchet to rectify thermal motion, though he proposed it purely as an interesting Gedankenexperiment, and did not analyze it.[7] Marian Smoluchowski analyzed in detail such a mechanism in 1914, where a linear ratchet is subject to thermal noise.[7] In Smoluchowski's proposal, a particle would more easily raise a ratchet than lower it, owing to the asymmetry of the teeth. However, if the pawl can be lifted by the fluctuations of the linear teeth, it could also be moved by the thermal noise itself—releasing the ratchet to freely lower and undo the work; with this process, no energy is ultimately gained, and the second law is saved. Feynman proposed instead a rotating ratchet, connected to a paddle wheel, which is subject to thermal fluctuations. Feynman showed that if the pawl and the paddle wheel are at the same temperature, the same thermal fluctuations rotating the wheel would occasionally release the pawl and undo the work. However, where the paddle wheel and the pawl are at two different temperatures, the ratchet can serve as a heat engine.[7] In other words, ratchet mechanisms can rectify nondirectional motion, but only if the system is kept away from equilibrium—some source of energy must be provided. Thermal noise can play a role in the motion, but cannot power it. The qualitative equation of a ratchet is: broken symmetries plus forces that are zero bias on average equal the directed current, in accordance with Curie's principle (a phenomenon will occur if not ruled out by symmetry).

In the mid-20th century, alongside these discussions in theoretical physics, chemists and biologists were struggling to understand the mechanisms powering biological motors. It was known that some proteins use chemical energy in the form of adenosine triphosphate (ATP) to produce unidirectional motion, but the specific processes and energetics were elusive. In some cases, they remain so, but such fascinating forays are far beyond the scope of this perspective.[8,9] In the 1990s, the connection between the motion of biological motors and the thermodynamic concept of ratchets came into focus.[10] The scale of motor proteins means that they must battle relatively powerful Brownian forces, of the same magnitude as those considered for Brownian ratchets.[11] Much like the asymmetry inherent in the gear, it was understood that biological motors employ asymmetric conformations and binding potentials. The ATP molecules consumed by the motors serve as the energy input, akin to the heat-engine nature of the thermodynamic ratchet.[7] Around the same time, experimental implementations of particle ratchets began to appear in the literature, with the earliest demonstrations transporting latex[12] and silica[13] microspheres using electric fields[12,13] and optical traps.[14]

PARTICLE RATCHETS: A CONCEPTUAL BRIDGE TO ELECTRON RATCHETS

The "Brownian motors" (so named because they appear to harness thermal noise to function) inside of proteins can be modeled as a particle moving in a one-dimensional potential. This analysis is particularly fruitful for motor proteins, such as kinesin, that walk along microtubules inside cells.[7,15] Here, the x-coordinate represents a state variable that characterizes the important geometrical configurations in the protein. The ratchet potential represents the asymmetric conformations and binding potentials, and the movement of the "particle" in energy along the y axis corresponds to the internal energy gain and loss from the hydrolysis of ATP and fluctuations from the cellular environment. This model can be used to test two hypotheses of the mechanism of Brownian motors: fluctuation (flashing) and power-stroke (tilting). A fluctuation-based motor pays ATP to determine when thermal noise has transiently granted it "free" motion along the desired direction. ATP is used to increase the energy of the "particle," i.e., reset the internal mechanisms of the motor, such that it can diffuse and explore the asymmetric potential landscape, which represents the internal asymmetry of the motor protein. Alternatively, the power-stroke motor transduces ATP to motion, by using the energy from hydrolysis to bias the potential energy landscape.

This concept can be extended to the ratcheting of particles with no internal structure. We can flip the source of energy and asymmetry from internal to external such that the ratchet potential and its modulation are externally applied as the particles move

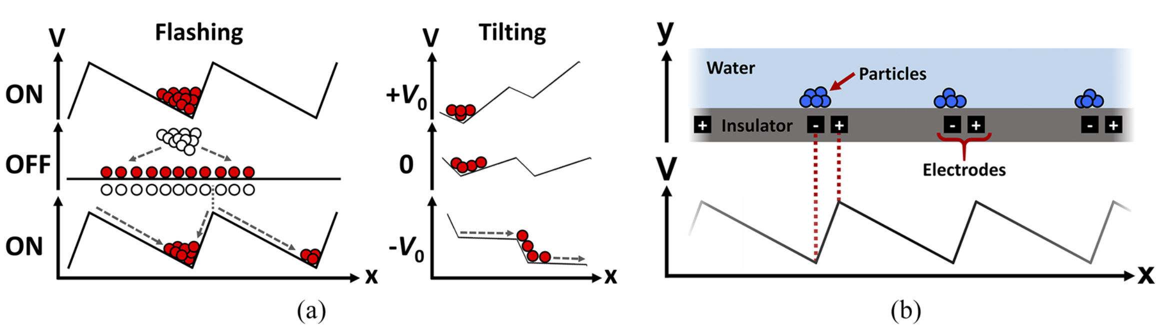

FIG. 1. (a) Operating principle of flashing and tilting ratchets. In a flashing ratchet, the magnitude of an asymmetric repeating potential is temporally oscillated, e.g., turned on and off, perpendicular to the direction of transport (here, left to right). No bias is present along the direction of transport. In a tilting ratchet, a bias is applied along the direction of transport, tilting the permanent asymmetric potential. (b) A common way of applying a flashing ratchet potential in experimental ratchets is to use pairs of oppositely charged electrodes under the transport layer. The asymmetric spacings of the electrodes allow the application of a sawtooth potential. Panel (b) reproduced with permission from Lau *et al.*, Mater. Horiz. **4**, 310 (2017). Copyright 2017 The Royal Society of Chemistry.

along a Cartesian coordinate. Figure 1 illustrates how flashing (previously fluctuating) and tilting (previously power-stroke) models can be applied to particles. The canonical ratchet has translation symmetry (is spatially periodic), while a pump only has one repeat unit.[16] We refer readers to reviews that discuss in depth different classes of ratchets, and their connection to Maxwell's demon.[7,16] The bridge from ratchets to electron ratchets is now conceptually simpler, as we only need to trade classical mechanics for quantum mechanics.

Ratchets are nonequilibrium systems and display complex behavior that depends on the time- and space-dependent flows of energy into and out of the system. The nonequilibrium flow of energy leads to the central defining feature of ratchets, *current reversals*, where the direction of particle current can switch upon the variation of some parameter.[15] Remarkably, if a current reversal is observed while varying one parameter of the system (e.g., AC frequency), it can also be induced by tuning any other parameter.[15] The ratchet current as a function of parameters is a high-dimensional space where current reversals can serve as the borders between different mechanisms of transport, with an unknown partitioning of the space between regions of parameter space that yield a finite current or zero current. A central focus of research in ratchets is the theoretical, computational, and experimental investigation of parameters that control current reversals. While it is possible to deduce the mechanism of a specific current reversal for a model or experimental system via careful analysis, there is no general theory that governs all current reversals. We will show in the remainder of this perspective that this knowledge is difficult to attain and transfer across fields because the complexity of ratchets introduces irreducible paradigms at any level of study.

THE FIRST ELECTRON RATCHETS

We continue our bridge from particle ratchets to electron ratchets through the study of the original literature that ushered in the field. In 1998, Song *et al.* studied the properties of a *geometric* rectifier based on the ballistic transport of electrons.[17] A triangular antidot was patterned into a 2D electron gas, and the device exhibited a nonlinear response, producing voltage of the same sign when the current changed sign, Fig. 2. The device could thus act as a full-wave rectifier. This device did not have a threshold voltage, unlike a typical semiconductor diode, and was proposed for the detection of weak signals. We emphasize that the symmetry of the antidot leads to the absence of current reversals, in accordance with Curie's principle.

In 1998, Linke *et al.* studied the transport properties of a quantum dot etched onto the surface of a 2D electron gas (GaAs/AlGaAs).[18] This time, the electron transport was done *through* the quantum dot, and the voltage drop along the dot, coupled with the quantization of energy levels, led to *nonsymmetric* resistance. Figure 3 plots the resistance vs voltage characteristics of the asymmetric quantum dot. A diode does not allow current (high resistance) until a certain threshold voltage, where it will abruptly switch on (zero to low resistance) and pass current up until saturation, determined by the material properties and internal geometry. Linke's quantum dot, however, exhibited different behavior. The lack of spatial inversion symmetry led to nonsymmetric resistance,

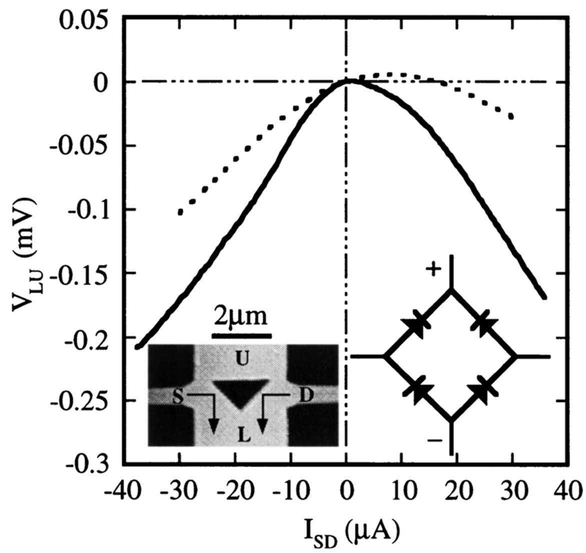

FIG. 2. The voltage developed (V_{LU}) vs the applied current (I_{SD}) for the geometric rectifier is shown in the left inset. The voltage developed is not symmetric vs the applied current because the symmetry of the device about the (LU) axis is not perfect. Reproduced with permission from Song *et al.*, Phys. Rev. Lett. **80**, 3831 (1998). Copyright 1998 American Physical Society.

where the measured resistance varied as a function of bias voltage and Fermi level. Thus, when tasked with rectifying an AC signal, a diode will produce a square waveform where the negative voltage portion of the signal is cut out, but the current produced by this quantum dot will highly depend on the geometry (which determines energy distribution of the states where transport occurs), the Fermi level, and the amplitude and frequency of the waveform.[19] This nonsymmetric resistance can lead to current reversals, which were observed in subsequent work that applied AC fields to a periodic array of such quantum dots.[19]

What is the microscopic origin of the nonsymmetric resistance? At the experimental temperatures (T = 0.3 K or 5 K), the electron mean free path due to impurity scattering is about 15 μm, much larger than the scale of the potential (1.7 μm), so the electrons can be treated as classical billiards. The asymmetry of the triangular quantum dot leads to different scattering rates in forward vs reverse bias, which can be shown from both a quantum and classical scattering formalism.[20] At T = 5 K, the results are explained purely by the ballistic motion of the electrons. At T = 0.3 K, however, fluctuations in the current emerge, which are related to the specific alignment of the density of states inside the dot, and to how the voltage drop is distributed over the contacts and the dot. Figure 4(a) shows how a periodic array of these quantum dots was used to rectify an applied AC voltage and generate a net current even in the absence of an average bias.[19] The net current is a *time average of the nonsymmetric resistance*, a static measurement. In fact, the time-dependent, zero-bias driving need not be a sinusoidal wave, as colored noise also leads to the ratchet effect.[10]

These papers were the first to describe a *rocking* (or *tilting*) electron ratchet—an AC bias in the direction of transport "tilts" the potential, but the time-averaged bias is zero. A rocking ratchet

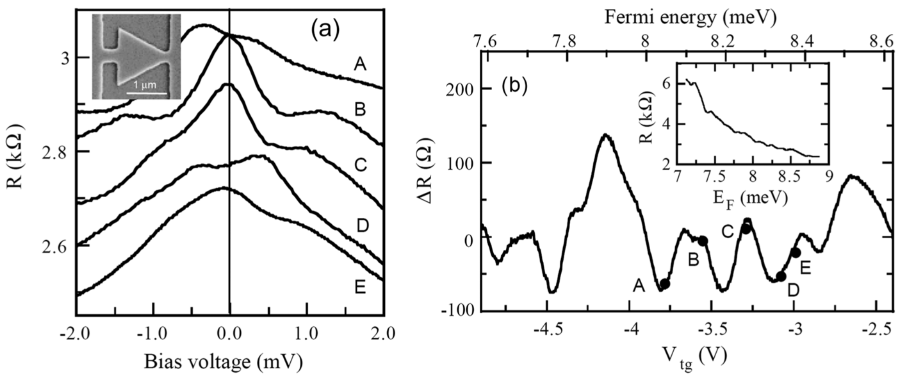

FIG. 3. (a) The measured resistance vs bias voltage for an asymmetric quantum dot, at various (ABCDE) gate voltages that control the Fermi level. Inset: a SEM image of the triangular quantum dot. (b) Resistance fluctuations at zero bias vs gate voltage. The gate voltage controls the alignment of the energy levels of the contacts with respect to the quantum dot states. At low bias voltages, the resistance vs bias curves in (a) correspond to traversing the resistance fluctuation curve in (b). Reproduced with permission from Linke *et al.*, Europhys. Lett. **44**, 341 (1998). Copyright 1998 EDP Sciences.

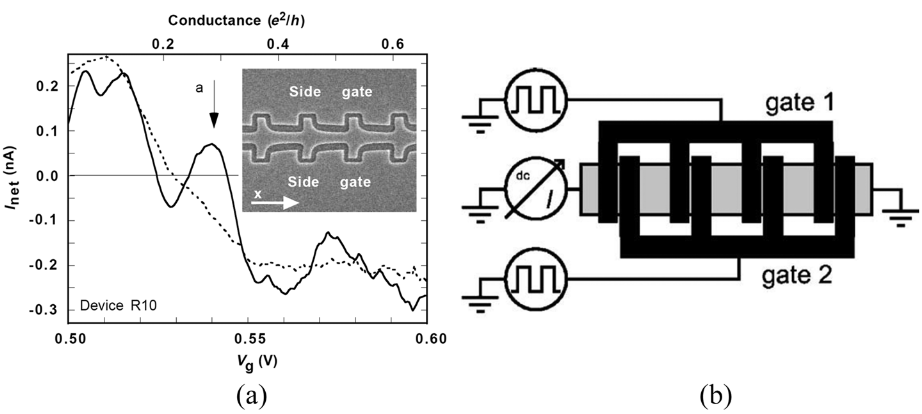

FIG. 4. Rocking and flashing electron ratchets. (a) Net current for a tilting ratchet at T = 0.4 K (solid line) and T = 4 K (dotted line). Inset: scanning electron micrograph of the device, showing four periods. Reproduced with permission from Linke *et al.*, Science **286**, 2314 (1999). Copyright 1999 American Association for the Advancement of Science. (b) A flashing electron ratchet, where the symmetry-breaking potential is applied with a pair of interdigitated electrodes. Reproduced with permission from Müller *et al.*, Appl. Phys. Lett. **87**, 042104 (2005). Copyright 2005 AIP Publishing LLC.

produces current as long as the electrons see the asymmetry of the potential during their ballistic free flight. Thus, the mean free path of the electron must be on the order of the asymmetry, and the mean scattering time also places an upper limit on the frequencies that the ratchet can respond to, analogous to dielectric freeze out. In practice, the time scale restriction means that a rocking electron ratchet can rectify time-dependent perturbations up to GHz (ns lifetime in typical semiconductors), but the length scale of the ratchet must be tuned to the mobility of the material. Increasing temperature diminishes the current in a rocking ratchet because the increase in kinetic energy allows the electron to spontaneously escape the barriers and also reduces the mean free path.

In contrast, a *flashing* ratchet modulates the amplitude of the potential itself (e.g., turning it on and off), without applying a momentary or net bias in the direction of transport. Figure 4(b) shows the first example of a flashing ratchet, where finger electrodes on top of GaAs/Al_xGa_{1-x} apply an asymmetric potential to electrons in the confined 2D electron gas.[21] The electrons alternate between directed and diffusive transport as the confining potential is turned on and off. Diffusive transport plays a central role in the operation of a flashing ratchet and is responsible for the different asymptotic frequency dependence of a rocking vs flashing ratchet—both types of ratchets cease to work in the high frequency limit, while a tilting ratchet can still function in the low frequency limit.

WHAT WOULD AN IDEAL RATCHET BE USEFUL FOR?

From this brief introduction, we now understand the qualitative fundamentals of a ratchet, and how they respond to different types of zero-bias driving (rocking and flashing). Similar to a rectifier, the two major suggested applications of electron ratchets are detection (sensing) and energy harvesting. In both cases, incident radiation is rectified to produce currents, but there are very different requirements and design choices.

In detection applications, we typically want to differentiate between different frequencies and different polarization states, and so we aim for highly tunable designs, to enable signal selectivity. A sharp frequency response would serve this end, assuming it can be tuned to select the signal of interest, either when producing the detector or, ideally, *in situ*. Additionally, the relationship between the intensity of the input and the output signal should be predictable, and stretch across a wide intensity range. Sharp intensity response curves (strong output changes for slight input changes) and measurable responses to weak signals are desirable; as is a device architecture that can be tuned to different frequency ranges, in fabrication or *in situ*. Successful sensors employ well-controlled structures—possibly with resonant elements—where the geometric shape and charge carrier mobilities of the device lead to strong current generation for a very limited set of conditions, or at least to sharp changes in the current as a function of signal properties.

In contrast, energy harvesters need to respond similarly to a wide range of frequencies and polarization state—current reversals must be limited to conditions far from any expected operating state. Furthermore, harvesters need to respond well to wide spectra, a combination of frequencies acting on the harvester simultaneously, rather than individual frequencies. Otherwise, one can imagine incoming radiation in a combination of frequencies (e.g., a solar spectrum) producing a variety of current magnitudes and directions, ultimately almost nullifying the net current. Instead, we seek broad and flat response curves, aside from cases of a well-characterized monochromatic energy source, such as for beaming power via single-wavelength radio waves.

The detection of THz-range radiation is a major challenge, and the ratchet effect was recently proposed as a solution. A 2015

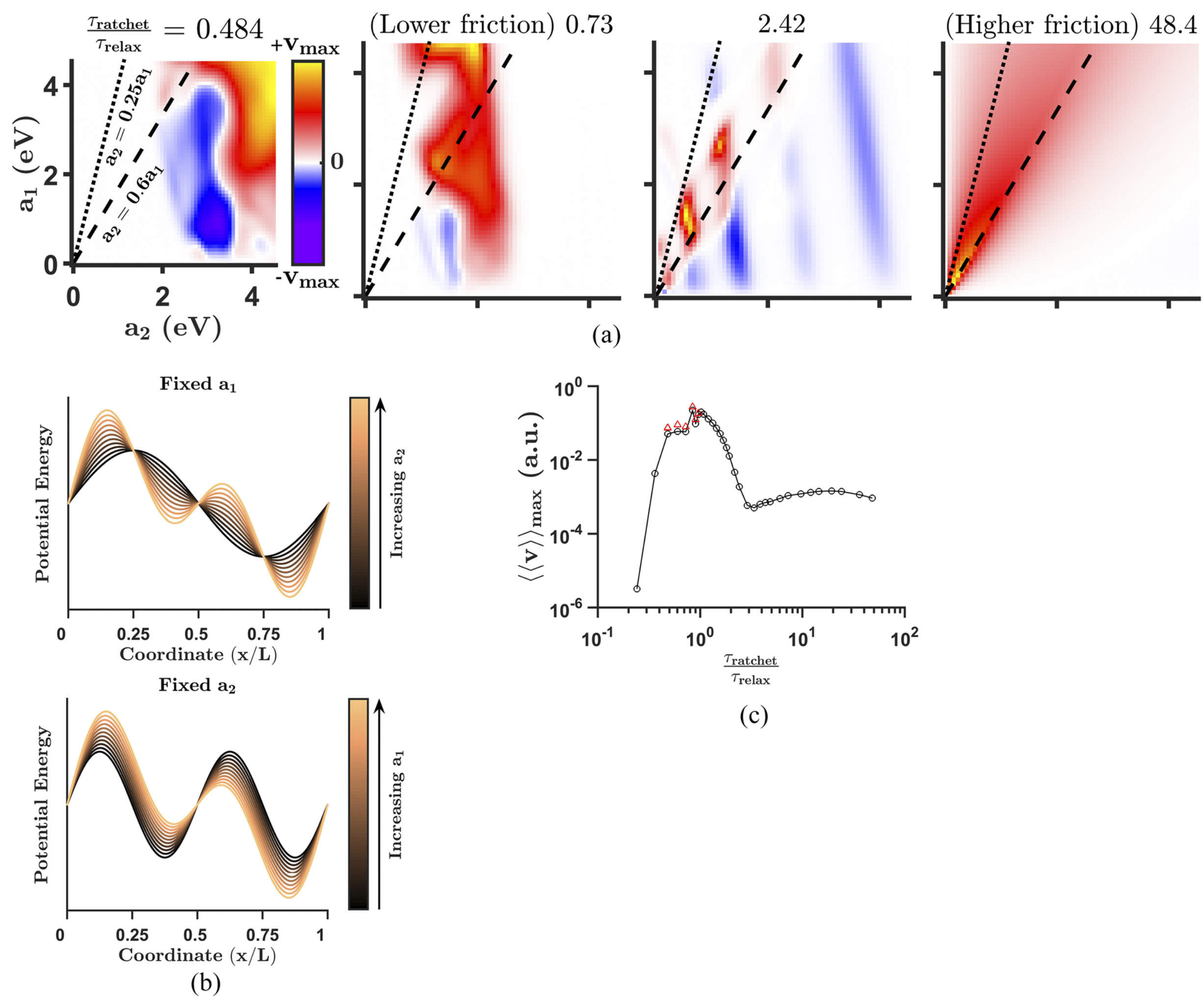

FIG. 5. (a) The normalized velocity (see shared color bar in the left-most plot) of a wavepacket traveling in a biharmonic potential under various oscillation times. Each pixel represents a different shape, determined by the values of a_1 and a_2 in the one-dimensional biharmonic potential $V(x) = a_1 \sin\left(\frac{2\pi x}{L}\right) + a_2 \sin\left(\frac{4\pi x}{L}\right)$; see the shared axis labels in the left-most plot; each of the four plots represents a different degree of friction, or damping, determined by the ratio of the driving time $\tau_{ratchet}$ and the relaxation time τ_{relax}. Multiple current reversals are observed in the low friction case for small modifications of the potential, but the response becomes smoother for increased damping. (b) Some of the potential shapes produced by varying a_1 and a_2. (c) The peak velocity, for any shape, as a function of the degree of friction. Adapted with permission from Lau *et al.*, Phys. Rev. E **93**, 062128 (2016). Copyright 2016 American Physical Society.

study by Faltermeier *et al.* demonstrated a ratchet-produced photocurrent, sensitive to the helicity of the radiation.[22] The studied device employed a high electron mobility transistor overlaid with a dual-grating gate, which provided the required asymmetry. A related work from 2016 by Olbirch *et al.* used an array of asymmetrically spaced metal bars as a grating or as electrodes on a graphene sheet, and also produced photovoltages for THz radiation.[23] The asymmetric grating (similar to the electrode pairs used in many flashing-ratchet studies) produces a similarly asymmetric electrostatic potential in the graphene and spatially modulates the applied THz electric field due to near-field diffraction. We have proposed a ratchet-based THz detector in a 2017 theory study, using strain gradients to set up a ratchet potential in silicon.[24]

The precise control and engineering of current reversals would be desirable in sensing, while the suppression of current reversals would be ideal in energy harvesting. Thus, understanding the mechanism of current reversals is critical to fabricating an electron ratchet for sensing or energy harvesting. We have shown in our 2016 computational work that the existence of current reversals is a quick test for the amount of damping (dissipation and friction) in the system—as damping increases, the response curve flattens.[25] In our system, the dissipation was implemented phenomenologically, while in real materials, electrons lose energy and coherence through phonon and impurity scattering. High dissipation increases the rate at which electrons relax to equilibrium, which in turn reduces the effects of asymmetry on the electrons through a reduction of their ballistic lifetime. Figure 5 shows that, for an underdamped system, the magnitude and direction of current strongly depend on the driving frequency and shape of the potential, while for an overdamped system, the current is far less sensitive to those parameters. Careful consideration of damping strength vs the characteristic time scale of electron motion will be helpful in future designs. The degree of damping in a system depends on the mobility of the electrons, the operating temperature, and the characteristic length scales.

WHAT HAVE WE LEARNED SINCE THEN? PROGRESS TOWARD FUNCTIONAL RATCHETS

To structure the following survey, we separate the studies by the type of ratchet device used, although for some devices, the categorization is somewhat arbitrary. We again note that we aim to discuss major developments based on our perspective of the field, using representative studies, rather than review each and every study.

Tilting ratchets

A study by Linke *et al.* in 1998 was followed the next year by one of a periodic ratchet potential, in the form of several quantum dots patterned on a surface, connected in series, as we mentioned above.[19] Operation of this tilting ratchet once again required cryogenic conditions, with reported operating temperatures of 0.4 K and 4 K. The direction of the rectified current varied with the temperature, and tunneling was shown to contribute to the flow of electrons. Since that original demonstration, this area has seen relatively little development, as the focus shifted to other ratchet designs. Very recently, Custer *et al.* entered the field with a tilting ratchet based on Si nanowires, fabricated to have asymmetric constrictions.[26] The constriction acts as a 3D geometric diode, where electrons moving quasi-ballistically are preferentially reflected in one direction, generating a current. The authors demonstrated room-temperature rectification up to an oscillation frequency of 40 GHz and showed that their devices differentiate between different modulations of a carrier signal. Commendably, the authors fabricated dozens of devices, to elucidate the relationship between structure and function.

Drift ratchets

In 1998, a related design for an electron ratchet was demonstrated, when Lorke *et al.* explored the behavior of an array of asymmetric antidots, Fig. 6.[27] Antidots are holes in a thin semiconductor film, which supports a 2D electron gas. Transport in antidot arrays was previously studied, but for arrays with reflection symmetry.[28,29] Here, the authors used aligned triangular antidots such that the array has no reflection symmetry, and this change allows it to produce a photovoltage. The low operating temperature (4.2 K) resulted in electron mean free paths much larger than the inter-hole distance, allowing electrons to move essentially ballistically. Far infrared radiation (119 μm) produced a photovoltage in the device, attributed to a ratchet effect. The limitation of cryogenic operating temperatures was quickly overcome in a 2001 study, which miniaturized the triangular holes to only 150 nm (side length), and the inter-hole spacing to 250 nm, both of roughly the same magnitude as the electron mean free path in the 2D electron gas at room temperature (136 nm).[30] Only some of the electrons travel ballistically under these conditions, and as expected, decreasing the temperature significantly enhanced the performance. The resulting device produced a DC voltage from an applied AC potential, up to 50 GHz. It

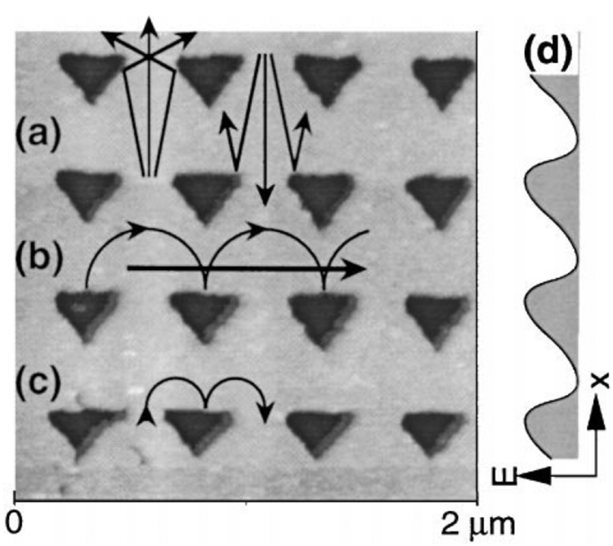

FIG. 6. (a)–(c) Atomic force micrograph of an antidot array etched in a GaAs–AlGaAs heterostructure, which supports a 2D electron gas, with possible electron trajectories. (d) A proposed equivalent 1D sawtooth potential. Reproduced with permission from Lorke *et al.*, Physica B **249-251**, 312 (1998). Copyright 1998 Elsevier Science B.V.

was noted by the authors that this frequency is already above the working frequencies that of most conventional diodes, and they expected the device to work for even higher frequencies. The antidot array design, whether using triangular or semicircular dots, was used in multiple other experimental demonstrations.[31,32]

Flashing ratchets

Though the flashing ratchet design had been the first to be experimentally demonstrated, those initially transported only microparticles and molecules, not electrons. This changed in 2005 when Müller *et al.* used two sets of interdigitated electrodes [see Fig. 3(b)] to impose a ratchet potential on a 2D electron gas, at T = 4 K and below.[21] When two phase-shifted oscillating signals were applied to the two sets of electrodes, a DC current was produced. The dependence of the current on the phase difference varied between the applied temporal waveforms (sine, rectangular, and triangle), and the results were reproduced by a hydromechanical model. Studies in 2011 and 2012 utilized a similar interdigitated electrode design to ratchet electrons in an organic semiconductor (pentacene) at room temperature and showed that the ratchet can produce power and do work against a bias.[33–35] Taking an important step toward useful applications of ratchets, the authors estimated the charge efficiency, that is, the net charge moved per cycle divided by the total charge moved (in either direction). The authors use a numerical model to estimate the overall power efficiency, but did not measure it experimentally.[35] With a combination of experiment and simulation, the authors showed that the peak frequency (the driving frequency producing the maximum current) increases with μL^{-2}, where μ is the charge carrier mobility, and L is the spatial period of the ratchet.[34] Despite the important advances, owing to the complexity of the devices, much of the observed behavior remained unexplained, including the current reversals, which the authors speculate were related to the presence of contacts breaking the symmetry in specific device configurations.[33]

The next advance in flashing electron ratchets came in 2013, when Tanaka *et al.* deposited asymmetrically shaped flat electrodes on a nanowire supporting a 2D electron gas.[36] Rather than relying on an array of electrode pairs to produce the asymmetric potential, here, the asymmetry of each individual electrode determines the potential. The ratchet produced a current at both room temperature and cryogenic conditions (T = 10 K). A 2015 follow-up work further explored the dependence on structural parameters of the ratchet.[37] Though a current reversal can be observed in one figure,[36] it is not commented upon.

In 2015, Mikhnenko *et al.* published a study of a single period ratchet, based on an organic semiconductor layer, operated in the flashing configuration.[38,39] This ratchet operates via a charge pump mechanism and rectifies an applied AC potential, which can be deterministic or stochastic (noise). The key to this device is the creation of a permanent asymmetry in the organic semiconductor P3HT [poly(3-hexylthiophene-2,5-diyl)], by first applying a high stress voltage (30–100 V), which asymmetrically distributes large ions in the polymer. The ion gradient imbues the semiconductor with a diode-like character. The authors later developed a lower-cost fabrication method,[40] used contacts of different work functions,[41] and created an n-type device, compared to the original p-type design.[42] These devices do not produce current reversals likely because they rely on the diode-like permanent asymmetry created by the stress voltage.

In 2017, we, together with Ratner and Weiss, introduced a different flashing electron ratchet design, which uses the asymmetric thickness profile of electrodes to apply an asymmetric electric potential to a light-responsive bulk-heterojunction transport layer [Fig. 7(a)].[43] The ratchets had a peaked dependence on the driving frequency, with large differences between the fabricated devices, including frequency-based current reversals [Fig. 7(b)]. Our 2016 theoretical study, discussed above, showed how sensitive the current direction and magnitude are to the shape of the potential,[25] and the desired (by design) and undesired (due to fabrication errors) differences in the shape of the electrodes are likely behind some of the variation. The ratchets were capable of doing work against a bias, and illumination with visible light modulated the current [Fig. 7(c)]; we measured an overall maximum efficiency of about 0.5%. Remarkably, the ratchet devices produced currents for temporally unbiased waveforms (having a zero time-averaged value, e.g., sine), whereas previous experimental implementations were limited to temporally biased functions, such as on/off, or $\sin^2$. We explored and explained this observation using further experiments with an adapted analytical model[44] and classical particle-based simulations.[45] The key lies in the 3D nature of the transport layer—the electric potential, applied using electrodes under the transport layer, decreases through the thickness of the layer. The trajectories taken by the charge carriers include movement in both the x- and z-directions, meaning that the thickness of the layer is critical to their behavior. Previous theoretical work only addressed 1D ratchet devices with piecewise linear sawtooth potentials, where ratchet transport for time-unbiased driving functions is prohibited.[46] This case shows how critical it can be to accurately capture the central features of experimental devices in theoretical models. The precise details of the physical implementation of the ratchet can have profound effects on its performance, and selecting which ones to include in a model is a non-trivial task.

A second fascinating observation in our experimental devices[43] was that although illumination increased the DC conductivity of the transport layer by photogenerating charge carriers, in many cases, it actually decreased the ratchet current, or even reversed its direction, an observation we ascribe to detrimental interparticle repulsion. In 2019, Kodaimati *et al.* explored this dependence in great detail, using light and solvent-annealing to vary both mobility and carrier concentrations.[47] With this rich dataset, the authors were able to describe the dependences on either parameter, as well as elucidate the impact of a higher level of asymmetry in the devices. In related work, Kedem and Weiss used further particle-based simulations to study the effect of interparticle repulsion on the performance of the ratchet, and uncovered two separate cooperative transport mechanisms, which increase the transport at high and low driving frequencies.[48] In the first mechanism, interparticle repulsion increases the effective diffusion rate of the particles and decreases the spatial periodicity, two effects which allow for operation at higher driving frequencies. In the second mechanism, transport occurs over a larger fraction of each oscillation, and through repulsive interactions, some particles gain enough momentum to traverse multiple spatial periods within a single oscillation.

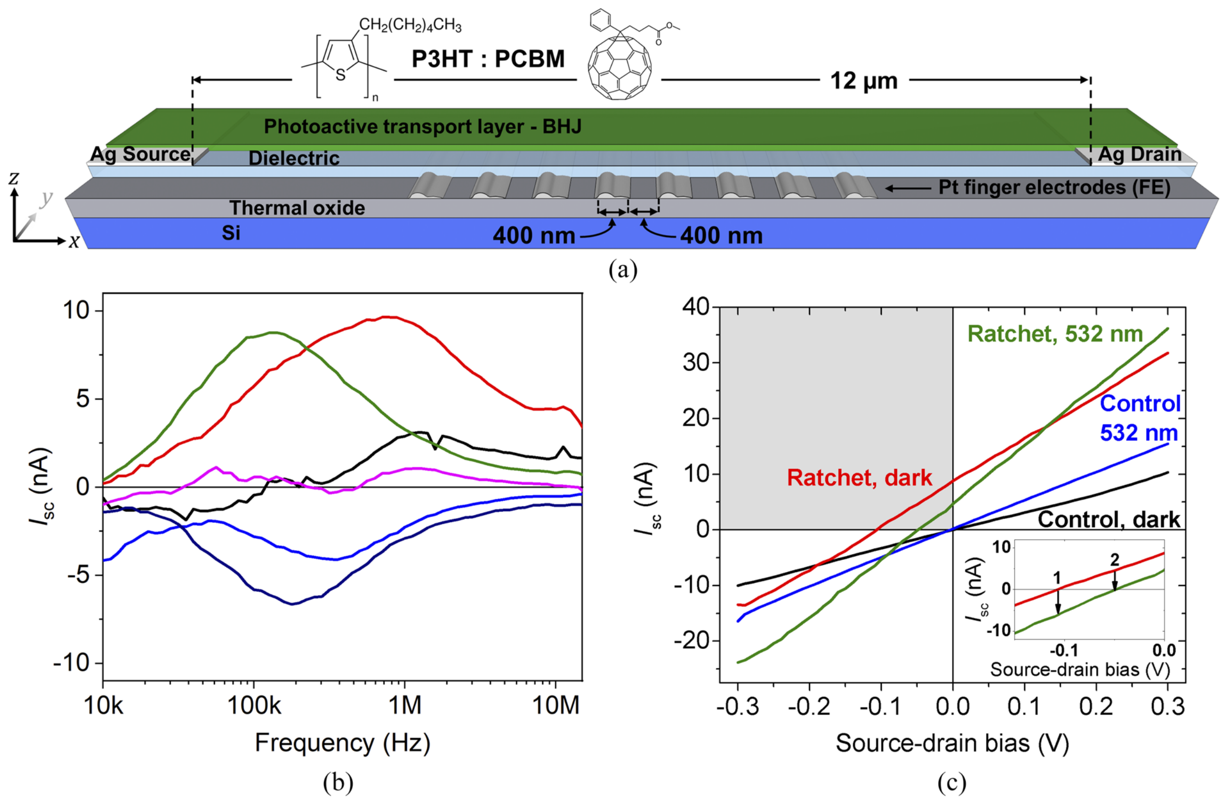

FIG. 7. (a) Schematic representation of the experimental ratchet device: eight Pt electrodes are embedded in a dielectric layer underneath a bulk heterojunction transport layer. An oscillating signal is applied to the Pt electrodes, and their asymmetric thickness profile induces an asymmetric potential in the transport layer, producing a current. (b) Short-circuit current for six flashing ratchet devices as a function of driving frequency. (c) Current vs voltage for one of the devices, with ("532 nm") and without ("dark") illumination, and with ("Ratchet") and without ("Control") the flashing potential. The ratchet is doing work in the upper left quadrant. Inset: a section of the *I*–*V* curves where illumination can turn the ratchet current on ("1") or off ("2"). Panels (a) and (c) reproduced with permission from Kedem *et al.*, Proc. Natl. Acad. Sci. U. S. A. **114**, 8698 (2017). Copyright 2017 authors.

The thread running through our studies is that the consideration of the fine details of experimental ratchets results in the uncovering of higher-level phenomena, which control the performance of the ratchets. As is common for non-equilibrium devices, small changes in the geometry or operating conditions produce large differences in the resulting behavior.

Seeking to progress toward useful devices, in a 2017 theoretical study, we, together with Kodaimati, Ratner, and Weiss, proposed a new design for a flashing ratchet. In the proposed design, individual electrons are excited above the level of the ratchet potential by interaction with incoming photons.[24] The potential is created by strain gradients in Si, and weak (below bandgap) excitations raise the energy of individual electrons enough to allow them to explore the potential surface for brief periods, Fig. 8. In essence, from the individual electrons' perspective, the potential is turned off upon excitation, until scattering events bleed the excess energy, and the electron relaxes to a potential well. Our simulated ratchet produced currents, which increased with the illumination intensity, and depended on the ratio of photon energy (in the ~10 THz range) to potential well depth. Though the predicted energy conversion efficiency was low, this was primarily due to computational limitations dictating non-optimal material choices, and we believe the design has potential for powering functional devices. Remarkably, the ratchet produced currents for below-bandgap, unpolarized, and incoherent radiation, a feat not shown before in any ratchet study. Interestingly, the ratchet showed no current reversals for any parameters studied, hypothesized to be due to the incoherent nature of light absorption.

Single-electron ratchets

In tilting, drift, and flashing ratchets, the asymmetric potential is either constant, or its magnitude manipulated uniformly, that is, the entire potential is increased or decreased everywhere, simultaneously. In a pump, however, parts of the potential are oscillated in time, and in combination with other features, allow or block transport in specific directions at different times. Electron pumps are sometimes termed single-electron (SE) ratchets and typically use three gate electrodes to define two asymmetric barriers and a central well; an alternating voltage is then applied to at least one of the

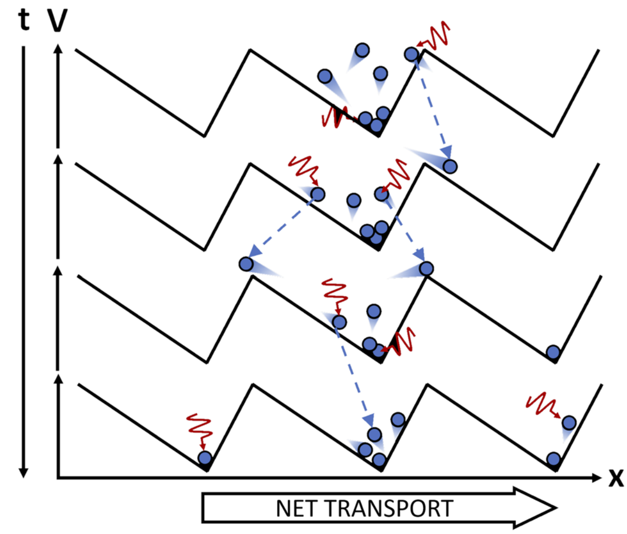

FIG. 8. In the proposed light-powered ratchet, below-bandgap photons (red) excite electrons (blue) in the conduction band of doped Si through intraband absorption. The electrons then explore the static potential through various elastic (impurity, acoustic phonon) and inelastic (optical phonon) scattering mechanisms. The dashed arrows schematically show various fates of electrons that are excited by intraband absorption. Reproduced with permission from Lau *et al.*, Adv. Energy Mater. **7**, 1701000 (2017). Copyright 2017 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

gates, to pump single electrons into and out of the well. Figure 9 shows the design and operating principle of a typical single electron ratchet. The year 2008 saw significant advances in this area, demonstrating DC currents from AC potentials in the GHz range.[49–51] In 2017, Chida *et al.* modified the design by adding a charge sensor to measure the occupancy of the single electron box, adding feedback capability, and making the device into an electronic Maxwell's demon.[52]

Symmetry breaking by magnetic fields

Ratcheting is predicated on breaking the inversion symmetry along the direction of transport. One way of doing so is to apply

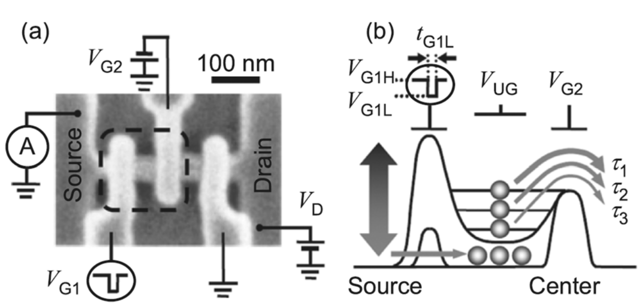

FIG. 9. (a) Scanning electron micrograph of a single electron (SE) ratchet. (b) Schematic representation of the area of the SE ratchet shown in the dashed frame in (a). Reproduced with permission from Miyamoto *et al.*, Appl. Phys. Lett. **93**, 222103 (2008). Copyright 2008 AIP Publishing LLC.

a perpendicular magnetic field. In a series of studies from 2013 onward, magnetic fields were applied to graphene,[53] a silicon MOSFET,[54] and quantum well structures overlaid with a dual grating.[55] The field provided the necessary asymmetry and allowed each of the structures to rectify the applied AC potential of an incident THz laser, and produce a DC photovoltage.

Domain-wall effect

Non-centrosymmetric crystals have long been known to produce a photovoltage, a phenomenon termed the bulk photovoltaic effect.[56] A potentially different effect was identified in 2010, when an above-bandgap photovoltage was measured in a ferroelectric device composed of $BiFeO_3$.[57] The authors propose an underlying mechanism involving sharp potential steps at domain walls (1–2 nm wide), which assist exciton separation. Though the connection to the ratchet field was not identified at the time, the proposed potential surface is qualitatively the same as the common piecewise linear ratchet potential, as shown in Fig. 10. Here, transport is produced due to the asymmetry between the sharp gradients at the walls (which lead to exciton separation), and the gradual gradients between them (which do not greatly affect excitons). As in the other ratchet systems described above, there is no overall potential gradient across the device, yet a photovoltage is obtained. The mechanism was further explored in later work, and the internal quantum efficiency was estimated at 10%.[58] Several lines of evidence support the authors' conclusion that this domain-wall effect is separate from the previously identified bulk photovoltaic effect, though this issue is still controversial.[59,60]

THE IDEAL VS REAL BEHAVIOR OF ELECTRON RATCHETS

The most abstract description of a ratchet is an equation of motion for a single particle in a spatially periodic potential, subject to time periodic (or colored noise) driving, with additional random thermal fluctuations. This formalism, both classical and quantum, can be qualitatively analyzed to determine the fundamental symmetries that must be broken for ratchet transport to manifest, as analyzed by Denisov *et al.*[61] First, the authors identified symmetry transformations that change the sign of an observable A, $A(t) = \mathcal{A}[\Psi(t)]$ but leave the equations of motion unchanged. Here, $\mathcal{A}$ is a functional that produces an observable from a trajectory $\Psi(t)$. Since the equations of motion are unchanged, the solution of the original equation of motion includes the trajectory that changes the sign of the observable. If these two trajectories contribute equally to the long-time observable, or are part of the same trajectory, then the expectation value of the observable is zero. Then, we can choose parameters of the potential and driving to destroy all symmetries identified in a system. This exhaustive analysis identified the spatial and temporal symmetries that needed to be broken for directed transport to occur for a variety of ratchet types (flashing, rocking, and traveling).

While we can understand the basic symmetry requirements for ratchet transport, we still do not know (1) the magnitude of the current, or location of current reversals, for any particular set of parameters for a ratchet, and (2) how a ratchet behaves when dissipation and noise are significant. Both of these questions are addressable

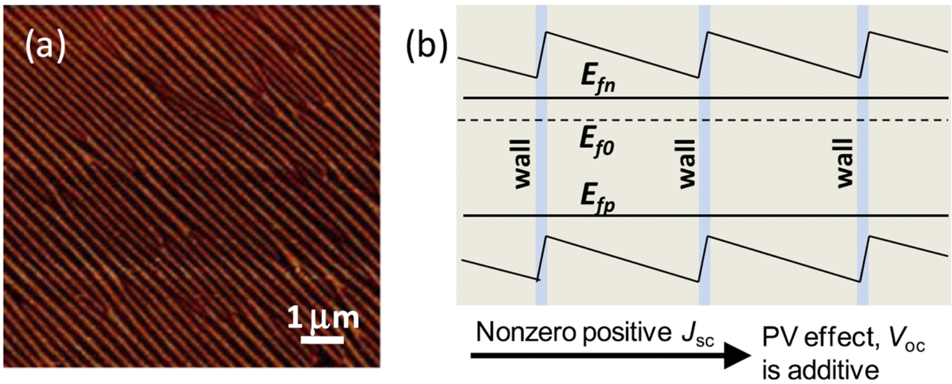

FIG. 10. (a) Piezoresponse force micrograph showing the domain walls in $BiFeO_3$. (b) Schematic representation of the short-circuit band alignment and current flow under illumination, showing the split quasi-Fermi levels E_{fn} and E_{fp} for nonequilibrium electrons and holes. Reproduced with permission from Seidel *et al.*, Phys. Rev. Lett. **107**, 126805 (2011). Copyright 2011 American Physical Society.

by numerical calculations. There is an extensive body of work that studies the parameter dependence of classical and quantum ratchets, specifically, a particle that is periodically kicked in time.[62–65] Figure 11 shows examples of the ratchet current as a function of dissipation and kicking strength. There is a great amount of structure in the system, with a family of shapes that are periodic in parameter space. Similarly, we[25] and others[66] have extensively explored the effect of shape on the ratchet current; however, we studied the effect of continuous, sinusoidal driving, while the other works use a delta-kicked potential. These exhaustive simulations have a sobering conclusion: there is a large disconnect between fundamental symmetry analysis, numerical simulations, and experimental realizations, with each separate body of work largely unable to inform the conclusions of the others. For example, a symmetry analysis cannot predict the existence of the periodic "shrimp" shapes [Fig. 11(a), orange, upper left quadrant] in the numerical simulations, and it would be difficult to experimentally realize a delta-kicked electron ratchet. Based on the numerical simulations, we can expect that small perturbations in experimental realizations of ratchets, due to systematic variations of parameters or even random error in synthesis and fabrication, can lead to large changes and reversal in the observed ratchet current. Strong dissipation and thermal noise will reduce but not wholly eliminate this parameter sensitivity, and come with the disadvantage of decreasing the maximum attainable ratchet current due to friction loss.[25]

Finally, interparticle interactions open up new modes of collective transport, as we discussed above (under *Flashing ratchets*), which represents yet another axis along the ratchet parameter space. There is no reason to expect the lessons that we have learned from exhaustive theoretical and experimental study of single electron ratchets to carry over to interacting electron ratchets, especially in the presence of strong correlation or electron–phonon coupling in actual materials.

REDUCTIONISM VS COMPLEXITY: EMBRACING A SYSTEM-LEVEL VIEW OF RATCHETS

There has been a great deal of effort made to understand current reversals in ratchets. At best, we can understand how to engineer them for specific systems, but we still cannot predict how they will occur when presented with a new, arbitrary system. This lack of understanding simply comes from the differences between an ideal and a real ratchet. The incredible sensitivity of the current reversal to variations in any parameter means that a model will almost always fail to capture the specific behavior of an experimental realization of a ratchet: deviations in shape, composition, applied fields, and importantly, *how* the ratchet responds to these deviations. There has been enough research done to firmly establish the fact that current reversals exist, but they are hard to control (see our discussion in the section "*The ideal vs real behavior of electron ratchets*"). Reductionism has dominated the bulk of research in ratchets, with a focus on finding basic principles through isolation and simplification, such as conducting experiments in cryogenic temperatures, manipulating atoms in optical traps, and studying single quantum dots and electrons. We believe that it is time to move from the study and modeling of idealized systems to a chemistry and materials science perspective, which embrace complexity and emergent properties, and adopt a system view of the scientific and engineering problem.[67] We suggest a pivot to a focus on identifying and realizing applications of ratchets, where theory aids in the design and optimization, taking into account synthesis, fabrication, materials, reproducibility, lifecycle, durability, and practicality.

As a case study, since the mid-1990s, there has been a persistent effort in applying the ratchet concept to photovoltaics in the form of an intermediate absorbing band.[68] The idea of adding extra absorbing states to a solar cell is an old one, originating from mechanically connecting solar cells of different bandgaps into a tandem cell. Each new bandgap added allows an additional wavelength of light to be absorbed, theoretically reaching up to 86% efficiency under concentrated solar light.[69] Tandem solar cells have been applied to many photovoltaic materials, from perovskites to organic to traditional inorganic cells. Figure 12 describes an intermediate band solar cell (IBSC), where this extra absorbing state is created within the host material.[70] The addition of an intermediate state makes an IBSC theoretically equivalent to a triple junction solar cell, but material properties and non-radiative recombination prevented an efficient realization of the IBSC.[70]

A solution to prevent non-radiative recombination is the use of a so-called ratchet band, a state lower in energy than the intermediate band, which does not allow transitions to the valence band. However, how is such a state to be realized? Figure 12(b)

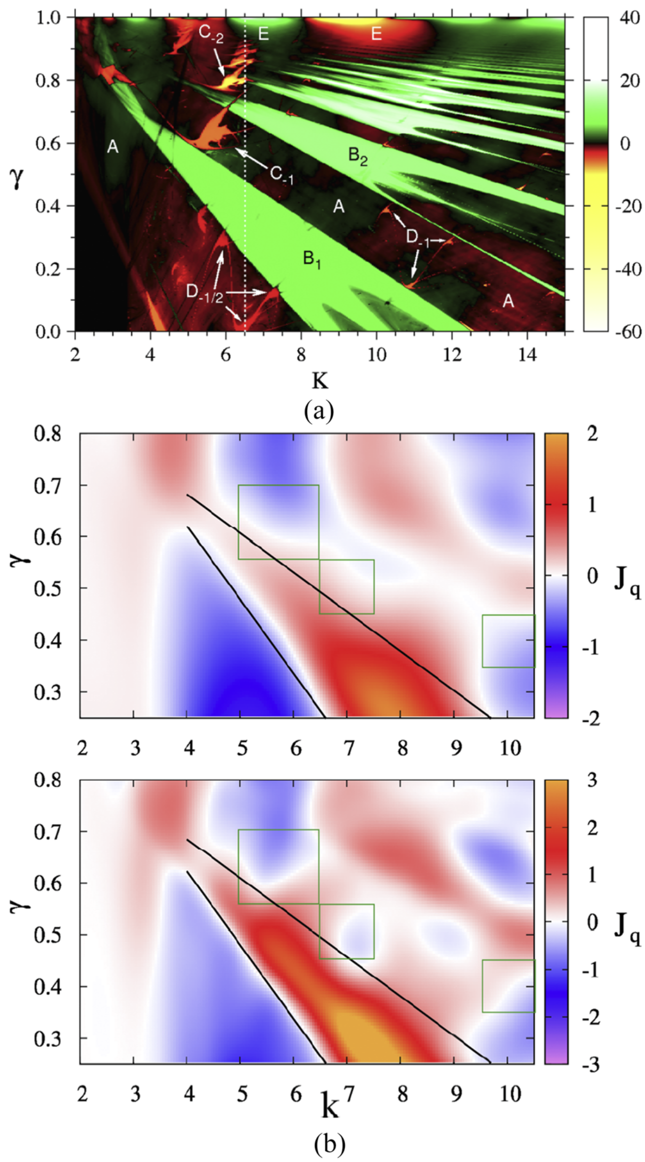

FIG. 11. (a) The ratchet current (dimensionless units) vs dissipation strength γ and kicking strength K for a classical ratchet. There are multiple shapes that appear in a periodic fashion as a function of the parameters, which are known to be generic structures in the study of dissipative maps. Reproduced with permission from Celestino *et al.*, Phys. Rev. Lett. **106**, 234101 (2011). Copyright 2011 American Physical Society. (b) The quantum current (dimensionless units) for the same model, where the top panel corresponds to a quantum solution and the bottom a more classical solution. Tunneling and coherence contribute to blurring the sharp lines that are seen in classical models. Reproduced with permission from L. Ermann and G. G. Carlo, Phys. Rev. E **91**, 010903 (2015). Copyright 2015 American Physical Society.

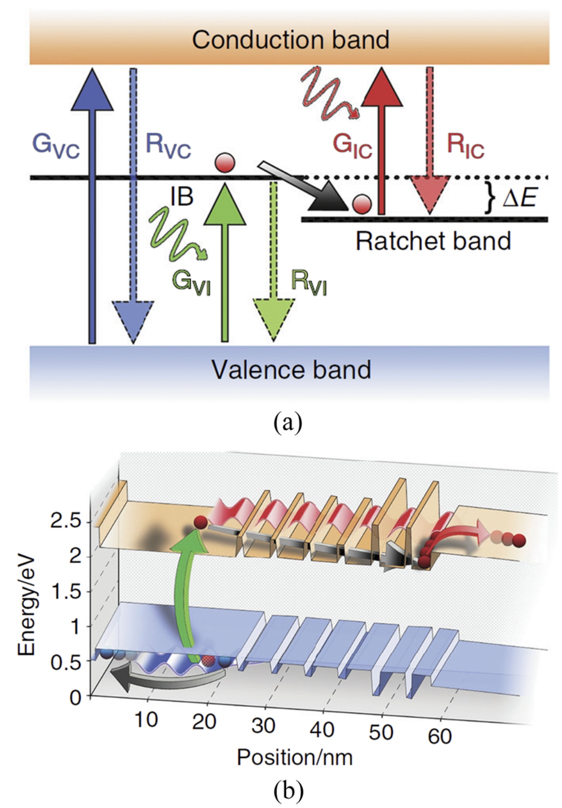

FIG. 12. (a) An energy diagram of an intermediate band solar cell with an irreversible transition (gray arrow) from the intermediate band to the ratchet band that prevents recombination from the intermediate to valence band. (b) A spatial view of the quantum ratchet. The ratchet band is realized with a quantum cascade that comprises a series of quantum wells: an excitation from a spatially localized hole state is quickly transported away through a quantum cascade to the ratchet band, where it is further excited into the conduction band. Reproduced from Vaquero-Stainer *et al.*, Commun. Phys. **1**, 7 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution 4.0 International License, http://creativecommons.org/licenses/by/4.0/.

depicts a spatial strategy where intermediate and valence band states spatially overlap, and the excited electron quickly moves away after the initial excitation to the ratchet band via a quantum cascade.[71] This source of asymmetry is exactly one period of a ratchet potential! A ratchet band solar cell is distinct from a highly mismatched alloy IBSC, where a swap between nitrogen and oxygen produces highly localized defect bands within the bandgap but still leaves excitations to the intermediate band vulnerable to nonradiative recombination.[72,73] Here, the ratchet band is spatially isolated from the intermediate band, which greatly reduces nonradiative recombination. The ratchet band solves two inefficiencies present in conventional solar cells, as it (1) allows the absorption of below-bandgap radiation and (2) provides directed transport for charge carrier collection instead of relying on diffusion. The theoretical work done for ratchet band solar cells was focused intensely on material considerations and bandgap engineering, which have mature knowledge bases and protocols in materials science, and directly translated to an experimental proof-of-concept ratchet band photovoltaic where the lifetime of an electron in the intermediate states is increased due to spatial separation.[74]

The role of theory in enabling this experimental demonstration was to aid in the design and optimization of the ratchet architecture.[75] This approach is important because there are material specific parameters, such as phonon scattering rates, that are only available with a material-based theory approach. For example, we have shown that optical phonon scattering is the most important mode of dissipation in controlling the efficiency of a silicon-based ratchet designed to capture terahertz radiation.[24] In contrast, theory in the physics community has largely focused on the exploration of model systems to discover fundamental quantum features of electron ratchets. The physics-focused and solar-cell focused ratchet fields have not experienced much cross-fertilization to date. A recent review of the IBSC literature mentions the keyword "ratchet" only three times and does not cite any physics papers discussing ratchets.[70] The optimization of ratchet band solar cells is almost entirely done with a steady-state, rate-based formalism, which could benefit from *ab initio* quantum chemistry for modeling the material properties and quantum cascades, and semiclassical and quantum dynamics for simulating the transport of electrons.

FUTURE DIRECTIONS

In reviewing the current state of the field, we identified several research directions that we believe hold significant promise to grow our understanding of electron ratchets, and bring us closer to functional ratchet devices:

Feedback

A central feature of biological ratchet mechanisms is feedback—the potential surface, which describes the possible conformations of the protein and its binding to other molecules, changes in response to events, such as binding and fuel consumption. Numerous theoretical treatments of particle ratchets have demonstrated significant benefits in power efficiency from using closed feedback loops, where the driving is adjusted based on the positions of the particles.[76–80] While this approach might be viable for ratchets transporting micrometer-scale particles, which can be observed using a microscope, it is impractical for electrons. In experimental electron ratchets, the charge distribution is unknown, and so the potential surface is oscillated [e.g., by incident electromagnetic (EM) radiation] without any consideration of the charge distribution. Furthermore, theoretical treatments generally neglect the effect of the charge distribution on the effective potential surface. As we mentioned above, Chida *et al.* recently built an electronic Maxwell's demon based on a single electron ratchet, where the occupancy of the electron trap could be measured, and barriers on either side of the trap raised and lowered to rectify the random motion of electrons.[52] This approach, however, will likely not scale to multi-electron ratchets, and still requires considerable external measurement and driving instruments, in contrast with the autonomous operation of biological motors.

A second, as-yet-unrealized approach would allow the charge distribution to tune the potential surface directly, to create an autonomous feedback circuit. This capability would greatly simplify the design of feedback-enabled ratchet, and increase their energy harvesting efficiency. This approach is particularly attractive for devices intended to be powered by electromagnetic radiation or other environment sources, and which cannot be tethered to measurement, analysis, and driving electronics. We have already seen evidence of this effect in our simulations,[48] as the interacting (classical) particles can screen the applied electric field and repel each other—giving rise to the collective behavior discussed below.

Collective effects

Aside from the unique case of single electron ratchets, multiple ratcheted particles can and do interact with one another. Owing to the high computational cost of tackling interparticle interactions, these are most often neglected in simulations, which thus correspond to the behavior at the low-concentration limit. Theoretical studies of 1D flashing ratchets with interacting particles revealed complex behavior, with increased, decreased, or even reversed current as the particle density rose.[81–86] Simulations of a drift ratchet[87] and a combined theoretical and experimental study of a tilting ratchet[88] show similarly complex behavior. We also recently observed the impact of increasing particle density in experimental work[43,47] and revealed two collective transport mechanisms in simulation work, discussed earlier.[48] Our simulation studies directly complemented the experiments as they allowed for tracking trajectories and the proposing mechanisms to explain the observed behaviors.

Even the limited set of conditions studied produced a wide range of behaviors, stressing the need for more systematic studies, and particularly ones exploring more realistic systems, where complex effects can emerge. Furthermore, none of the above theoretical studies explicitly treated electrons, but rather studied idealized particles, or outright nanoparticles, in largely uniform environments. How would the unique behaviors of electrons manifest in a multi-electron simulation? The answer to this question is crucial for a deeper understanding of practical electron ratchets.

Flexible experimental platforms

We found that the greatest impediment to the systematic study of electron ratchets is the small range of parameters we are able to study, owing to the inflexibility of experimental platforms. By and large, experimental electron ratchets are produced using microfabrication techniques, and most or all of their structural parameters are fixed at the time of fabrication. Driving parameters, such as potential amplitude and frequency, can be varied over a wide range, but the basic structure of the ratchet and crucially the asymmetry in the potential are fixed. Lacking the ability to produce thousands of finely controlled devices, we simply cannot efficiently explore the wide parameter space. As ratchets can have very sharp dependence on multiple parameters, generalizations are difficult, limited, and often unwarranted. For these reasons, the development of an easily reconfigurable ratchet would be a boon for studies in this field. Rather than rely on fabricated electrode arrays to provide the asymmetry, such a platform would allow for changing the asymmetry on the fly, or with only minimal effort.

Complex and multifrequency driving

Experimental and theoretical ratchet studies to date have utilized simple driving waveforms (sine, square, etc.), at individual frequencies. However, if we learn more about the trajectories of electrons during each oscillation cycle, could we not modify those individual steps, by tuning the applied waveform? Perhaps, the rise of the square wave should be sharp, but the fall—slow. Perhaps, each cycle should contain finely controlled individual "kicks" to corral the electrons to the optimal path. Furthermore, in some cases, e.g., energy harvesting, the driving waveforms might contain multiple frequencies (even multitudinous frequencies, such as in the solar spectrum). How would those impact the behavior of ratchets, vs well known single-frequency driving?

Stiff vs sloppy parameters

All of the directions we propose are yet more axes in the high dimensional ratchet parameter space. The influence of any one parameter on the ratchet current is unknown and could be disentangled by identifying "stiff" and "sloppy" combinations of parameters. A change in stiff parameters leads to large changes in the ratchet current, while changes in sloppy parameters lead to little to no changes in the ratchet current.[89] This concept can be visualized as an n-dimensional skewed polygon. The coordinate axes that make up a skewed direction have high linear dependence, as movements along the skewed direction can be expressed as many possible combinations of those axes, i.e., the axes represent sloppy parameters. In particular, the effects of a set of sloppy variables can be exchanged for one (stiff) parameter, which is exactly the case when tuning current reversals. A stiff–sloppy parameter analysis could help guide the design and fabrication of ratchets, especially in the design of experiments as fabrication can be time-consuming and difficult to replicate, as well as identify structure in large-scale numerical studies of ratchet current.

SUMMARY AND OUTLOOK

We propose that ratchets can be used for sensing and energy harvesting, but we also need to take careful note of the competition. Research in THz detection is a very active field, from optical to material-based solutions.[90] Similarly, research in photovoltaics is a vast and fast-moving field, even in the subfield of intermediate band solar cells. What kind of efficiency can we expect from an ideal ratchet? Theoretical estimations of efficiency in molecular motors at maximum power range from 50% to near 100%, where the unity efficiency corresponds to a reversible, infinitely slow process (and hence no power).[91] Experimental measurements of the power conversion efficiency of biological motors range from 20% (kinesin)[92] to near 100% (ATP synthase),[93] where the wide range may be related to the reversibility of the process and their function—kinesin is a walker protein attached to a tubule, where directionality (thus irreversibility) is central to its function of transporting payloads across the cell, while ATP synthase rotates in one direction to generate ATP, and the other direction is powered by ATP. Thus, we can expect the efficiency of a ratchet to be affected by the depth of the potential wells relative to the thermal noise, i.e., how reversible is the transport mechanism. Should ratchets be developed as an add-on to enhance the functions of current technology, e.g., to enhance charge separation and collection in the existing photovoltaics, or should they be the primary mechanism of sensing and energy harvesting? The maximum attainable efficiency for electron ratchets is still unknown and is very likely dependent on the specific experimental realization, as we have elaborated on extensively in the body of this work. The ratchet band solar community uses the possible gains in efficiency to motivate their work, and we suggest that theoretical estimations of efficiency should play a strong guiding role in future research as well.

As the ratchet field continues to develop, it is important to remain cognizant of symmetry laws and the second law of thermodynamics. Ratchet transport often arises in quite unpredictable circumstances and sometimes produces only weak signals. Under these conditions, it is easy to mistake an instrumental or computational error for a true signal. While performing experiments or simulations, one must always ask oneself: How does this system actually break inversion symmetry? Where is the energy from transport coming from? If those questions cannot be satisfactorily answered, the observed signals might be spurious.

In summary, we reviewed the state of the field of electron ratchets. Basic research has laid down broad foundations, but there are not many attempts to build upon them. The sensitivity of the current to changes in any parameter makes it hard to translate lessons from the existing research to applications. We draw hope from studies of efficiency in molecular machines—biological ratchets—that highly efficient and highly tunable electron ratchets can find use in sensing and photovoltaic applications.

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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