NANOSCALE RECTENNAS WITH SHARP TIPS FOR ABSORPTION AND RECTIFICATION OF OPTICAL RADIATION

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1. INTRODUCTION

One of the major challenges in the efficient detection and conversion of energy in the electromagnetic spectrum is the development of a broadband device that will collect and rectify electromagnetic radiation from the IR through the visible portion of the spectrum, extending to $10^{15}$ Hz and higher. Current MIM and silicon based Schottky diodes are narrow band devices, rectifying up to the mid IR range of the electromagnetic spectrum ($\leq 10^{14}$ Hz).

We present a method for optical rectification that has been demonstrated both theoretically and experimentally and can be used for the development of a practical rectification device for the electromagnetic spectrum including the visible portion. This technique for optical frequency rectification is based, not on conventional material or temperature asymmetry as used in MIM or Schottky diodes, but on a purely geometric property of the antenna tip or other sharp edges that may be incorporated on patch antennas. This “tip” or edge in conjunction with a collector anode providing connection to the external circuit constitutes a tunnel junction. Because such devices act as both the absorber of the incident radiation and the rectifier, they are referred to as “rectennas.” Using current nanofabrication techniques and the selective Atomic Layer Deposition (ALD) process, junctions of 1 nm can be fabricated, which allow for rectification of frequencies up to the blue portion of the spectrum (see Section 2).

In rectennas, incident radiation is directed to a receiving antenna with one edge terminated by a tip or sharp edged structure that is part of a geometrically asymmetric metal vacuum/metal tunnel junction (with a gap distance $s$). The selection of an appropriate structure
such as a patch antenna, whisker antenna, or rectenna with a sharp edge coating, including diamond, BN, GaN, AlN, AlGaN, plasmonic materials, and Cs. is determined by the application, and its ability to absorb the incident radiation. The coatings reduce the magnitude of the forward tunneling barrier allowing for enhanced electron emission and rectification. In addition to the nano-geometry of whisker antennas, other realizations use patch antennas, extended solid and open geometries (e.g. squares, rectangles, any n-sided structure or others) and can operate into the IR and higher frequencies. The geometric parameters for such antennas are matched to resonance for a narrow band device and, for wide band devices, the geometric parameters are chosen based on energy absorption and energy density. The gap distance is designed so that the tunneling time is sufficiently short for electrons to transit the barrier before field reversal.

Due to the incident radiation AC currents are induced along the length of the antenna, which produce oscillating charges at the top or edge of the geometrically asymmetric tunneling junction and corresponding image currents in the anode. The presence of the constricted geometry of the tip or edge gives rise to an enhanced field at the tip. The oscillating charges in the tunnel junction induce an AC voltage across the gap. If the induced field is sufficient for field emission, a tunneling current is produced. Due to geometric asymmetry (and possible material asymmetry or plasmonic coatings), there is a difference between the potential barriers for forward and reverse bias, which results in a rectified DC current.

The specific type of geometrically-asymmetric device that we use in our own research is described in Section 4 and is shown in Figs. 13, 14, and 15. Note that we have chosen a device can be fabricated on a large scale, referred to as a Monolithic Nanoscopic Tunnel Junction (MNTJ).

The extension of rectennas from the microwave to the IR and visible regimes offers enormous potential benefits. In addition to the breakthroughs in our understanding of basic physics, optical rectennas would be useful in many transformative applications, including photovoltaics (the conversion of photon energy to electrical energy), solar cells (the conversion of solar energy to electrical, thermal or chemical energy), nano-photonics, near field optics, IR sensing, and imaging (including medical and chemical sensors). Another increasingly important application is the transmission and reception of information. This is significant since the density of transmitted information is greater at higher frequencies, where the density varies as the square of the frequency. Furthermore, for transmission through the atmosphere, losses decrease as the frequency increases.\textsuperscript{1,2}

Perhaps in direct proportion to the potential benefits, optical rectification has faced important challenges in materials processing and theoretical understanding. As the result of recent advances in nanotechnology, Metal/Vacuum/Metal (MVM) tunnel junction gaps can now be reproducibly fabricated down to \( \sim 1 \) nm over \( \text{cm}^2 \) sized areas using selective ALD. In particular, for Cu, the selective ALD process is self-limiting at gap separations of 1 nm. For gap distances of this size, rectification of radiation with frequencies in the visible range is possible. The theoretical understanding of the operation and description of antennas at the nanoscale in the optical regime is only now being studied in a rigorous way taking into account that the behavior of metals in the optical regime, which differs from that at frequencies below the IR.

In this chapter, we survey the results of our modeling, characterization, and nanofabrication of a geometrically-asymmetric rectenna device that acts as both an antenna and rectifier for IR and optical radiation. In Section 2, we review the response time of such devices, focusing on the results and implications of an important study by Nguyen et al. In this section, we explain how tunnel junctions are capable of rectifying signals in the visible regime. In
Section 3, we review the mechanisms of rectification, experimental data confirming optical rectification, and, finally, quantum-based theoretical analyses. In Section 4, we present a number of operational designs for the fabrication of optical rectennas. Section 5 gives a brief summary and an outline of plans for future work.

2. RESPONSE TIME OF TUNNEL JUNCTIONS

In addition to the issues regarding the fabrication of reproducible nanoscale devices, the response time of the rectifying device to optical radiation is a critical element for successful operation. The response time consists of several contributions. One is the collective response of the conduction electrons that establish the AC bias. Generally for metals, the collective response corresponds to frequencies well beyond the UV (or periods of about $10^{-16}$ sec). Two other elements affecting device response time are the electrodynamic response of the junction to the changing fields (RC-time) and the “transversal time” for electrons to cross the gap region in the tunnel junction before field reversal. These latter two times are considered in the following subsections.

2.A. “Traversal Time” or “Tunneling Time” for Nanoscale Tunneling Junctions

The concept of “traversal time” applied to electron transmission through time-dependent barriers, is needed to estimate the limiting frequency for the tunneling rectifiers used in nanoscale devices. Qualitatively, an electron of a given energy incident on the oscillating barrier “interacts” with the barrier for a time, $\tau_b$. Consider the two limiting cases. In one limit where the period of the oscillation, $T$, of the radiation is longer than this time of interaction, the electron effectively interacts with a “static” barrier and, hence, can tunnel before the field direction reverses. On the other hand for the limit where the frequency of the radiation is very high with $\tau_b << T$, then the electron interacts with many cycles of the radiation and the tunneling barrier is essentially unchanged due to the oscillating voltage. In this limit, the tunneling current is comprised of the photo-excited electrons which have absorbed or emitted quanta equal to $n\hbar\omega$, where $n=1,2,...$ and $\omega$ is the angular frequency of the incident radiation. The crossover between these two limiting behaviors may be determined by the relationship, $\omega\tau \approx 1$. The validity of such a conceptual approach has been the subject of debate and controversy ever since the advent of quantum mechanics and the recognition that there can be particle tunneling through classically forbidden barrier regions. Basically the problem lies in the difficulty of defining and measuring the traversal time for the simple time-dependent scattering experiment in which an electron represented by a wave packet tunnels through a spatially localized barrier and is detected beyond the tunneling region.\textsuperscript{4,5,6} A seminal experiment by Nguyen et al.\textsuperscript{7} used a dynamical approach to probe tunneling times in which a natural time scale is provided by a laser that is an integral part of the experimental arrangement. The laser incident upon an STM junction, consisting of a W-sharp tip and a polished, flat Si (111) anode, causes the tunneling and, at the same time, provides a “clock” to measure the duration of the event. Given that the laser induced electric field is larger near the pointed apex of the tip than at the planar surface of the sample means that the vacuum tunnel barrier will tend to buckle inward (concave) or become thinner for forward bias and balloon outward (convex) or become thicker for reverse bias (see discussion and Figure 5 in Section 3.A). Moreover, if there is material asymmetry as in the Nguyen STM junction, there is an additional barrier asymmetry introduced. Such an STM junction can be a rectifier and under irradiation leads to a net DC current.
It can be argued that, if for a fixed spacing the laser frequency is too high, few electrons will be able to transfer from one electrode to the other during the half of the period when the electric field vector in the laser beam accelerates the tunneling electron. This means that one should observe a cutoff in the strength of the rectified DC signal either 1) when the frequency is increased beyond a critical value while maintaining the tip-to-surface distance \( s \) fixed or 2) when the gap width \( s \) is increased beyond a characteristic value \( s_c \), while keeping the laser frequency constant. This latter method was used in these experiments, when the junction was illuminated by a 1.06-\( \mu \)m YAG laser. The tip-to-base gap \( s \) was then progressively increased until the laser-induced current vanished. The DC rectified current as a function of gap width for fixed frequency indicated a cutoff distance of about 2.5 nm for the 1.06 \( \mu \)m YAG laser line.

The Nguyen study explained such experimental results in terms of a simple model that assumes that the particle acts as if it obeys the kinematical equations of motion as the particle traverses the classically inaccessible region defining the barrier at a velocity approximately equal to the Fermi velocity. If we assume an average tunneling velocity to be the Fermi velocity, \( v_F \), then \( f_{\text{cutoff}} = \frac{v_F}{s} \). This analysis predicts that for a 1 nm gap with a metallic tip and vacuum barrier, the transit time of about \( 10^{-15} \) seconds corresponds to radiation approaching the UV.\(^7\) The technological difficulty of producing arrays of nanometer gap junctions over areas of cm\(^2\) has recently been overcome by Gupta and Willis using selective ALD.\(^8\) Planar arrays of Cu-vacuum-Cu tunnel junctions were produced on silicon wafers using conventional lithography techniques, followed by selective ALD to yield tunnel junctions of \( \sim 1 \)nm (Figs. 5 and 6.) This selective atomic layer deposition (ALD) process that is self-limiting at gap separations of 1 nm for Cu. At this spacing, the tunneling time is sufficiently short for electrons to transit the barrier before field reversal in the visible frequency range, leading to rectification for asymmetric barriers.

These estimates for the “traversal” time have been corroborated in a series of simulations by Mayer et al.,\(^9,10,11,12,13\), who have used a quantum-mechanical transfer matrix approach for the
modeling of a geometrically-asymmetric, metal-vacuum-metal junction subject to an oscillating potential. This quantum mechanical scheme accounts for the three-dimensional aspects of the problem as well as the time dependence of the barrier. The currents are obtained by solving the time-dependent Schrödinger equation with a Floquet expansion of the wave function. For simulations using a full range of frequencies in the solar spectrum, Mayer et al. investigated how the efficiency of the rectification is affected by the aspect ratio of the tip, the work function of the metallic elements and the occurrence of polarization resonances. Their results demonstrate that the rectification of infrared and optical radiation is possible using devices of the type considered in this review.

2.B. RC-Time for Geometrically-Asymmetric Tunneling Junction (GATJ) Rectifiers

For the case of a planar MIM structure, the RC response-time of the junction is limited by parasitic capacitance yielding a practical limit of 10-100 THz.\textsuperscript{14} By contrast, point-contact devices (i.e., whisker diodes, and GATJs whose geometry is essentially the same) have been used in measurements of absolute frequencies up to the green part of the visible spectrum, demonstrating a response time of the order of femtoseconds, orders of magnitude faster than conventional MIM diodes.\textsuperscript{15} The asymmetrical, non-planar geometry of the pointed whisker in conjunction with the flat anode is an essential requirement for increasing the cutoff frequency $\omega_c$ of the diode, but inconsistent with the planar geometry of MIM tunneling theory for which the cutoff frequency is independent of contact area. In earlier studies of the detection and harmonic generation in the submillimeter wavelength region, Dees\textsuperscript{16} emphasized the importance of using the point-contact geometry to reduce the shunting effect of the capacitance and thus increase the high frequency cutoff of the device. Indeed the response time $\tau = 1/\omega_c = RC$ is independent of contact area for a planar MIM geometry since $C$, the capacitance of the contact, is proportional to $A$, the contact area, whereas $R$ the resistance is inversely proportional to $A$. On the other hand, for a point contact geometry, it can be shown using a solvable model with a spherical tip that $\omega_c$ is no longer independent of the tip radius (or area), and the sharper the tip, the faster the response time of the diode.\textsuperscript{14,17} Although mechanical stability of these earlier devices placed a limitation on producing robust sharp tips, modern fabrication techniques have overcome the mechanical fragility of previous point contact diodes and issues related to reproducible fabrication of nanoscale devices.

Below we provide a more detailed discussion of why it is necessary to use GATJs with sharp tips to obtain RC times short enough for rectifying radiation at high frequencies.

Such models are important for understanding individual device operation and final integration of devices into complex circuits. These device-circuit equivalents, such as that for a tunneling diode, allow for the direct application of the Kirchhoff Current and Voltage Laws which are, in turn, applications of conservation of charge and energy. In determining the equivalent device model, resistances ($R$), capacitances ($C$) and inductances ($L$) are used either as lumped (wavelength independent) or distributed (wavelength dependent, transmission line, $R$, $L$, $C$ per unit length) elements.

For the case of the rectenna with a GATJ rectifier, a lumped circuit model consists of a resistance for the metallic antenna taking into account its geometric properties (i.e., the tip that is part of the GATJ) while the junction is modeled as a capacitance with a large shunt resistance. This junction corresponds to the traditional modeling of a low leakage capacitor. Such a junction and resistive line feeding element represent a single time constant circuit as illustrated in Fig. 3. It can be shown that this transient circuit has a time constant $\tau$ given by:
\[ \tau = RC \left( \frac{R_s}{R + R_s} \right) \]  

(1)

Fig. 3. Equivalent circuit model of a rectenna with metallic antenna resistance \( R \) and junction capacitance \( C \) in parallel with junction shunt resistance \( R_s \)

The lumped circuit element \( R \) in Fig. 2 includes implicitly the geometrical nature of the circuit elements. It should be noted that except for the pointed antenna tip coupled to the junction anode (not vacuum), the quantities \( R^{-1} \), \( C \), and \( R_s^{-1} \) are all proportional to \( A \). However, for an antenna modeled as a spherical tip, the antenna resistance \( R \) is proportional to \( r_s^{-1} \), the inverse of radius of curvature of the tip. Hence, the response time \( RC \) is proportional to \( r_s \) or \( A^{1/2} \). The smaller the tip radius, the shorter the response time.

Alternatively we can consider the same circuit but analyze its steady-state behavior under an ac signal rather than under transient conditions. Following Sullivan et al.\(^\text{17}\), we now define \( \omega_c \) as the frequency at which half the power is dissipated in the series resistance \( R \). For this traditional half-maximum power limit for operation, the result is:

\[ R = \frac{R_s}{1 + \omega_c^2 C^2 R_s^2} . \]  

(2)

For \( \omega_c^2 C^2 R^2 \gg 1 \), this leads to the following condition for \( \omega_c \):

\[ \omega_c^2 C^2 R_s R = 1 \]  

(3)

We now demonstrate that \( \omega_c \) is proportional to \( r_s^{-1/2} \). The quantity \( C \) is proportional to \( A \), \( R_s \) is proportional to \( A \), and \( R \) is inversely proportional to \( r_s \); hence,

\[ \omega_c = \sqrt{\frac{1}{R_s RC^2}} \propto \frac{1}{\sqrt{(1/\pi r_s^2) r_s^{-1} (\pi r_s^2)^2}} \propto r_s^{-1/2} \]  

(4)
As the tip radius decreases the cutoff frequency increases. This reasoning led to the use of “ultra-fine” tips in the absolute frequency measurements of Javan and collaborators\(^{18}\) in which a thin, Tungsten wire several microns in diameter and approximate length of 1 mm was mounted at the end of a coaxial cable. The tip of the W wire was sharpened by means of a standard etching technique to a diameter of less than 100 nm.

Although neither the transient nor ac circuit approaches truly represent the GATJ but represent reasonable models for the rectifier, these approaches indicate that the RC response of the GATJ depends on the radius of curvature of the tip or contact area. The RC time constant has a dependence ranging between being proportional to \(r_s\) and \(r_s^{1/2}\), thus predicting that sharp tips used in the GATJ rectifier can be used to produce devices that can rectify radiation in the visible region.

As an example, we estimate the RC time constant of a rectenna with a GATJ (coupled to the anode) device with geometric parameters associated with one of our prototype devices described in Section 4 and illustrated in Fig. 14. We consider a typical periodic unit cell of this device, with a single nanoantenna. The prototype device consists of ten thousand unit cells, placed in parallel. For this first-order estimation of the RC time constant, we use material parameters that correspond to the limit when \(\omega \to 0\). It is understood that frequency-dependent parameters should be used when considering frequencies in the visible, in particular for frequencies at which polarization resonances occur. The experimental device is essentially a two-dimensional flat structure. The periodic unit cell in our modeling has a length \(L_x = 350 \text{ nm}\) along the \(x\)-axis and a cathode-anode spacing \(L_y = 295 \text{ nm}\) along \(y\). A value of 100 nm is taken for the thickness \(W\) of the structure. We represent the antenna by a flat triangle whose apex is replaced by a half-circular disc of radius \(r_{\text{apex}}\) that connects smoothly to the sides of the triangle. Initially, before the ALD metallization, the antenna has a base \(B = 110 \text{ nm}\), a height \(H = 245 \text{ nm}\) and a radius of curvature \(r_{\text{apex}} = 16 \text{ nm}\). The gap spacing between the apex of the antenna and the anode is 50 nm. The Ohmic resistance is estimated from a simple model in which we represent the antenna by the succession of slabs. For Cu, we obtain an Ohmic resistance \(R = 0.737 \Omega\).

In order to determine the geometric capacitance \(C\) of a unit cell of the device, we calculate numerically the electrostatic energy \(CV^2/2\), under a static voltage \(V_{\text{ac}}\) between the cathode and the anode. The electric potential in the unit cell of the device is obtained by solving Laplace’s equation \(\nabla (\varepsilon \nabla V) = 0\) (this expression is relevant to the static limit in which \(\omega \to 0\) and the antenna is assumed to have a dielectric constant \(\varepsilon \to -\infty\)). The resolution of Laplace’s equation is achieved by using a finite-difference technique.\(^{19}\) This resolution provides the electric potential \(V(r)\) in the system from which we can compute the electric field, \(E = -\nabla V\). The geometrical capacitance \(C\) of the system considered is finally obtained from the relation

\[
\frac{1}{2}C V^2 = \iiint_{\text{unit cell}} \frac{1}{2} \varepsilon |E|^2 \, dv
\]  

(5)

in which the right-hand side provides the electrostatic energy one must provide to a unit cell of the device in order to establish \(V_{\text{ac}}\). For the system represented in Fig. 14, we obtain a geometric capacitance \(C = 2.94 \times 10^{-18} \text{ F}\) and a resulting RC time constant of \(2.2 \times 10^{-18} \text{ s}\), a value corresponding to frequencies beyond the visible.
We can study how the parameters $R$, $C$ and $\tau = RC$ are modified by a conformal two-dimensional expansion of the antenna. A series of antennas are generated for which the parameters $B$, $H$ and $r_{\text{apex}}$ used for Fig. 3 are multiplied by a common dilatation factor $\alpha$ where $\alpha$ ranges from 1 to 1.204. In this expansion, the height $H$ of the antennas increases progressively from 245 nm to 294 nm, so that the gap spacing between the apex of the antenna and the anode is reduced progressively from 50 nm to 1 nm. The base $B$ of then increases at the same time from 110 nm to 132.45 nm. The parameters $L_x$, $L_y$ and $W$ of the device are kept constant. The Ohmic resistance $R$ of the antenna remains approximately constant. The geometric capacitance $C$ ranges from $2.94 \times 10^{-18} \ F$ (for $H = 245 \ nm$) to $1.69 \times 10^{-17} \ F$ (for $H = 294 \ nm$) and the RC time constant increases from $2.2 \times 10^{-18} \ s$ to $1.2 \times 10^{-17} \ s$, respectively. This dependence of the capacitance $C$ as $H$ increases from 245 nm to 294 nm is represented in Fig. 4.

![Capacitance C of a unit cell of the device, when the antenna is affected by a conformal planar expansion with the dilation parameter goes from 1 to 1.204. The results are represented as a function of the gap spacing d between the apex of the antenna and the anode, as H increases from 245 nm to 294 nm and the radius of curvature increases from 16 nm to 19.3 nm. The Ohmic resistance R of the antennas including the sharp tip is calculated to be 0.737Ω.](image)

From the simulations, the RC time constant of the device structures in this work should not be a limiting factor for applications related to the rectification of optical radiation, where the static values of the parameters are applicable. The extension of these simulations taking into account the frequency dependence of the material parameters is currently under investigation.

3. RECTIFICATION IN TUNNEL JUNCTIONS

3.A. Mechanisms for Rectification in Tunnel Junctions

The I-V characteristics of a tunneling junction are determined by 1) the flux of electrons in given initial states incident on the barrier interface and their occupation probabilities that depend on temperature and field, 2) the available final states and their occupation probabilities, and 3) the shape of the tunnel barrier, which may be modified due to contact potentials, surface photovoltage effects, induced AC voltages due to laser irradiation, etc. Correspondingly, the current asymmetry (or rectification properties) at fixed gap width $s$ must originate from one or several possible causes discussed below, namely material, geometrical, thermal asymmetry, and...
photo-stimulated changes in the electron flux distribution.\cite{20,21,22} We describe here only the geometric asymmetry of the rectenna device that is the critical element for achieving high frequency rectification.

For nanometer gap distances, the nature of the tunneling phenomenon is such that the current passes predominantly through that sharp protrusion closest to the planar sample surface. In such conditions and even in the absence of any material asymmetry (e.g., W tip, W surface, and assuming no work function inhomogeneities), the shape of the tunnel barrier is asymmetric as a function of the applied bias field. This is due to the geometric asymmetry of the electrodes comprising the tunnel junctions. This effect is illustrated in Fig. 5.

![Figure 5](image)

**Fig. 5.** (c) A tunnel barrier of a materially-symmetric, geometrically-asymmetric junction. The barrier exhibits a concave shape for (a) forward bias and (b) convex shape for reverse bias.\cite{7}

It is evident that the static electric field gradient is larger near the pointed apex of the tip than at the planar surface of the sample. This means that the vacuum tunnel barrier will tend to buckle inward or become thinner for forward bias and balloon outward or become thicker for reverse bias. The first observation of the geometrical asymmetry effect in a STM was observed by Feenstra et al.\cite{23} and in the detailed study of rectification in an STM presented by Nguyen et al.\cite{7} In addition, Dagenais et al.\cite{24} have experimentally verified that a geometrically asymmetric tunneling diode can be used to rectify radiation through the RF region. Based on their experiments, they envision that higher conversion efficiencies will be achieved at mid IR frequencies. Most recently, Ward et al.\cite{25} have shown both experimentally and theoretically that nonlinear tunneling conduction between gold electrodes separated by a subnanometer gap leads to optical rectification, producing a DC photocurrent when the gap is irradiated by a 785 nm laser source. In a recent paper, the authors have reviewed experiments verifying the geometrical rectification mechanism.\cite{26} Extensive simulations studies of geometrically asymmetric tunnel junctions verify these observations. In particular, the work of Mayer et al. provides insight into the development and optimization of devices that could be used for the efficient energy conversion of infrared and optical radiations. The studies also demonstrate that an accurate treatment of nanoscale tunnel junctions operating in the near IR and visible requires a quantum-mechanical treatment.\cite{9-13}
3.B. Experiments Verifying Rectification Mechanisms from the Microwave through the Visible Region

In this section we review some of the experiments verifying that an STM and other nano-junctions structures can act as antennas and rectifying devices for electromagnetic radiation from the microwave through the visible.

In a series of experiments by Kuk et al.\textsuperscript{27,28} used an STM consisting of a metal tip (Au) and a semiconductor sample (including Si(111)−(7×7)) was illuminated with laser radiation below and above the semiconductor indirect band gaps, specifically, photon energies of 2.94 eV, 1.96 eV, 1.17 eV, and 0.95 eV. The STMs were modified using a small lens to focus the laser beam to near its diffraction limit, yielding power densities up to 5.0 kW/cm\(^2\) on the junction.\textsuperscript{29} The resulting induced bias was measured laterally along the surface as light-induced excess current and voltage. For photon energies exceeding the band gap energy, surface photovoltages (SPV) of about 300 mV were induced across the gap independent of illumination intensity and frequency. For a photon energy of 0.95 eV, no surface photovoltage was detected. A small, atomically scaled (laterally along the surface) varying dc signal of 3 to 5 mV was also observed in the experiments. The authors suggest that this small signal is due to optical rectification associated with the geometric asymmetry of the junction. These striking results using a Au tip and collector demonstrate that the STM junction can absorb and rectify radiation corresponding to wavelengths shorter than 1.06 µm in agreement with the experimental results of Nguyen et al.\textsuperscript{7}

Tu et al.\textsuperscript{30} have experimentally verified that an STM junction can rectify radiation in the microwave region, which has led to the first direct, quantitative measurement of the rectification current due to single atoms and molecules. In their work, microwave of known amplitude and frequency irradiated the junction of a low temperature scanning tunneling microscope producing an electric field between the tip and an atom or molecule on the anode surface. It induced a DC signal that is spatially localized and exhibits chemical sensitivity at the atomic scale.

In 1998, Bragas et al.\textsuperscript{31} used a laser with wavelength of 670 nm to irradiate an STM junction to determine the field enhancement as measured by optical rectification. A field enhancement factor between 1000 and 2000 was obtained for highly oriented pyrolytic graphite and between 300 and 600 for gold. Analysis of their data indicated optical rectification due to junction geometry as well as thermal asymmetry. The admixture of \(\frac{\partial^2 I_{\text{stat}}}{\partial V^2}_{V_{\text{dc}}}\) was determined to be significant only for p-polarized light and in phase with the intensity variation, consistent with the expected behavior for the rectified current. Their experiments indicate that visible light (640 nm) can be rectified using nm-sized tunnel junction devices.

Most recently, Ward et al. have shown both experimentally and theoretically that “nonlinear tunneling conduction” between gold electrodes separated by a subnanometer gap leads to optical rectification when the gap is irradiated by a 785 nm laser source, producing a DC current.\textsuperscript{25}

3.C. Simulation Studies of Geometrically Asymmetric Tunnel Junctions

Unlike a conventional planar MIM diodes, devices employing a pointed nanowire tip achieve rectification solely or primarily with geometrical asymmetry.\textsuperscript{32,33} This is illustrated in Fig. 6 where the rectification ratio of a W-W junction (no material asymmetry) is compared using planar and pointed geometries. Figure 6 shows clearly that a planar geometry (with \(r \to \infty\)) provides no rectification, i.e., a rectification ratio (forward current divided by reverse current) of one.\textsuperscript{32}
Early theoretical work on the tunneling characteristics of these junctions usually relied on approximations in the shape of the barrier and in the tunneling probabilities, which were typically based on a one-dimensional model. However, modern computational facilities make it possible to address this problem more rigorously using quantum-mechanical techniques including three-dimensional aspects of the detailed atomic structure and the tunneling barrier. Such studies make it possible to investigate the geometric, material, and operational parameters that are important in optimizing the performance of geometrically-asymmetric devices.

Lucas et al. used a formulation of elastic, one-electron tunneling through three-dimensional, non-separable, spatially-localized barriers within the context of potential-scattering theory. They applied this approach to a model metal-vacuum-metal junction, consisting of two parallel electrodes, one of which containing a hemispherical protrusion. The electronic structure of each metal electrode is assumed to be free-electron-like. They found that the current distribution peaks within a narrow angle around the boss axis, confirming earlier estimates based on transfer-Hamiltonian formalism and in agreement with the observed atomic resolution of an STM microscope, when operating with atomic-size tips.

Mayer et al. presented a transfer-matrix analysis of a GATJ with a flat anode and a cathode with a hemispherical protrusion. This work confirmed the conclusions of Lucas et al. and explored how the rectification properties of such systems depend on their physical and geometrical parameters. This analysis still relied on a quasi-static approximation, in which it is assumed that one can compare currents obtained for static values of the external bias. This approximation is valid in the far-infrared ($\omega \rightarrow 0$) but must be replaced by a more exact approach in order to treat situations in which the time that electrons take to cross the junction is comparable with the period of the oscillating barrier.

In a subsequent paper Mayer et al. extended their previous work by taking into account the time dependence of the external bias explicitly using the transfer-matrix approach and the time dependent Schrödinger Equation. They assume that the geometrically asymmetric tunnel junction consisted of a cathode metal supporting a hemispherical protrusion with a height of 1 nm, a radius of 0.5 nm, and separation between the apex of the tip and the planar electrode of 1 nm. Due to the external electromagnetic radiation of varying frequency and intensity, there is an
impressed oscillating potential across the junction, \( V(t) = V_{ac} \cos(\omega t) \). In the simulations, \( V_{ac} \) varies from 0.01V to 1.0V and frequencies that correspond to quanta of energy between 0.2 eV (\( \lambda = 6200 \text{nm} \)) in the IR and 5eV (\( \lambda = 248 \text{nm} \)) in the UV. The rectification ratio that one obtains by taking the ratio of the mean values of the forward, \( I^+ \), and reverse, \( I^- \), currents is plotted in Fig. 7. The values obtained at low frequency, \( \omega \to 0 \), agree with those obtained in the quasi-static analysis.\(^9\) Because of the photon-absorption processes, the rectification ratio, \( R \), first increases with \( \omega \) before decreasing at higher frequencies. The intermediate region proves that the rectification of optical frequencies can be achieved by the device, which agrees with conclusions reached earlier by Sullivan, et al.\(^{14}\) In a quasi-static analysis, they predict a cutoff of the rectification for a photon-energy around 4 eV (radiation with wavelength of 300 nm in ultraviolet) because the field would then reverse before the electrons can cross the junction. Indeed, this oscillating-barrier analysis shows a significant decrease in rectification at that frequency.

![Fig. 7. Rectification ratio as obtained for a geometrically asymmetric junction subject to an external bias \( V_{ac} \cos(\omega t) \) with \( V_{ac} = 1 \) (solid line), 0.1 (dashed) and 0.01 V (dot-dashed line). The quantum of energy \( \hbar \omega \) ranges between 0.2 and 5 eV. The vertical lines indicate the height of the surface barrier (as measured from the Fermi level of the emitting metal) when \( V_{ac} \) (dashed line, left), −1 (dashed line, right), and 0 V (solid line).\(^{10}\)](image)

In order to assess the importance of the deposition of noble metals, known to have plasmonic resonances, on the field enhancement and rectification properties of a model tunneling junction, Mayer et al. performed 3D quantum mechanical computer simulations of optically irradiated MVM tunnel junctions using Ag and W tips.\(^{11}\) They predict an enhanced rectification and current output due to the surface plasmonic resonances in Ag at ~3 eV, corresponding to the energetic green portion of the visible spectrum. This study also explained the role of these plasmons and more generally of the frequency-dependence of the dielectric function on the
rectification properties of the junction. In Fig. 8, we plot the results of their simulations. Compared to tungsten, the power gained by the electrons that cross the device, and the rectification ratio of the device are enhanced by several orders of magnitude at frequencies that correspond to a resonant polarization of the tip.

These results suggest that the dependence of the plasmon frequencies on both the material and the geometry of the tip could be used to control the frequency at which the junction is the most efficient for the rectification of external signals. It is also important to note that there is a significant contribution of multi-photon processes (especially for $\hbar \omega \approx |eV_{ac}|$), which is most pronounced when a polarization resonance occurs. This effect opens the possibility to build devices for the selective detection of radiation in the infrared or visible domain or for a more efficient rectification and conversion of their energy. It is expected that deposition of a thin layer of other noble metals on an underlying antenna structure such as tungsten, molybdenum or aluminum should yield similar results.

![Rectification ratio of a junction made of silver and subject to an external bias $V_{ac} \cos(\omega t)$, with $V_{ac}=1$, 0.1 and 0.01 V (downwards, as indicated). The solid, dashed and dot-dashed lines indicate results using $\varepsilon(\omega) \rightarrow -\infty$, $\varepsilon(\omega) = 1 - \omega^2 / \omega^2$, and $\varepsilon(\omega) = 1 - \omega^2 / \left( \omega^2 + i\omega / \tau \right)$, respectively, as models for the dielectric function of the tip. Results obtained at the 3.1 eV resonance energy with $\varepsilon(\omega) = 1 - \omega^2 / \omega^2$ are off-scale and not shown.](image)

Mayer et al. also observed a significant enhancement of the energy conversion at frequencies that correspond to a resonant polarization of the tip. The dependence of these resonance frequencies on the shape and on the material used for the tip therefore gives the possibility of controlling the frequency at which the device is most efficient for the rectification of external radiation. It also was shown that reducing the work function of the metallic elements increases the performance. For practical applications, one may consider two-dimensional arrays in which devices would be placed with a typical spacing of 10 $\mu$m between adjacent protrusions. The currents and energies achieved per square meter correspond in this case to those achieved...
for a single tip times a typical factor of $10^{10}$. These results demonstrate that the rectification of radiation with typical frequencies in the infrared and optical domains can be achieved by using geometrically asymmetric, metal–vacuum–metal junctions. The results also provide a more quantitative analysis of the efficiency with which the energy of incident radiation can be converted by such devices.

The work of Mayer et al. provides insight into the development and optimization of devices that could be used for the efficient energy conversion of infrared and optical radiations. The studies also demonstrate that an accurate treatment of nanoscale tunnel junctions operating in the near IR and visible requires a quantum-mechanical treatment.\textsuperscript{9-13}

4. OPERATIONAL DESIGNS FOR AN OPTICAL RECTENNA

As stressed above, the operational design of a device that can harvest and rectify radiation from the infrared to the visible relies on antenna-coupled, fast tunnel diodes that employ geometrical asymmetry to realize efficient rectification. Given the evidence that the point-contact STM geometry makes this possible, one would like to nanofabricate nanostructures that are monolithic analogs of an STM junction.

One concept of a rectenna based on this novel approach consists of nanowires/mCNTs on planar substrates with a point contact-like rectifying junction as shown in Figure 9. The device uses wavelength-dependent-sized vertical arrays of nanostructures with point-contact junctions. The nanowire or mCNT forms a MVM or MIM junction barrier. As suggested, fast charge transport via tunneling is possible since capacitance is drastically reduced (over conventional planar junctions) and junction response times are on the order of a femtosecond.\textsuperscript{17}

Scaling such a rectenna device to production requires sufficient control of the geometric structure to achieve uniform and reproducible asymmetry, with the aim to manufacture large arrays of junctions with the necessary gap dimensions on the order of a nanometer. In order to achieve this level of control, Willis et. al., have developed a method for making scalable, nanofabricated tunnel junctions using atomic layer deposition (ALD). ALD is a thin-film growth technique that provides conformal deposition with enhanced control of the film growth rate for nanofabrication of devices with critical dimensions of a few nanometers or less. This ALD process provides sub-monolayer precision of the growth increment with average growth rates near 0.05 nm per ALD cycle. Such precision is typical of ALD growth and has led to numerous applications such as nanometer thin films for transistor gate oxides and highly dispersed nanoparticles for catalysis.

As outlined in Fig. 10, the combination of this ALD method with conventional nanofabrication techniques allows for robust tunnel junction nanofabrication. A nanotemplate seed-layer with the desired structural features is made using conventional nanofabrication techniques. Subsequently, the wafer with nanotemplates is loaded into an ALD reactor and metal is deposited onto the template structures via ALD growth. The layer-by-layer growth
causes the electrodes to converge toward tunneling with submonolayer precision. Typically, a few representative devices are monitored in-situ and the process is completed when the devices achieve tunneling with the desired gap resistance. Fig. 11 illustrates typical electrical data recorded in-situ during growth. Note the change in tunneling characteristics with increasing ALD cycles.

**FIG. 10.** Schematic of nanoelectrode fabrication and measurement. (a)-(b) A metallic template layer is converged to tunneling by selective ALD. (c) Device array. (d) In-situ electrical data showing convergence to tunneling and self-limiting effect (i.e. saturation).
There are two critical aspects of the process: The first is selective growth, i.e. selective ALD, which occurs on the nanotemplates and not on the insulating regions surrounding the devices (typically SiO$_2$ or Si$_3$N$_4$). Selective ALD is required so that as-fabricated devices are electrically isolated and there are no leakage currents; as a result, only a tunneling current is measured between electrodes. The second critical aspect is that tunnel junction growth is self-limiting. The devices do not short-circuit with excessive numbers of ALD cycles. Rather, the devices reach a self-limiting nanogap near 1 nm due to molecular scale transport limitations of the relatively large ALD precursor – Cu(tmhd)$_2$ – with a molecular diameter near 1.2 nm. The major accomplishment has been to control these critical aspects in order to achieve working tunnel devices. Modifications and enhancements of the above-described process include: selective growth on one, but not the other, of a nanoelectrode pair; variations of the seed layer; and scale-up to devices working in parallel.$^{35,36}$

![Fig. 11. Tunneling current vs. applied voltage for select numbers of ALD cycles.](image_url)
Combining e-beam and conventional lithography with selective ALD enables fabrication of the idealized thin-wire structure shown in Figure 12 (a) – (c). The plasmon layer is deposited using selected area ALD of copper which selectively deposits Cu only on the antenna and electrodes. The self-limiting growth process can create a gap between the collector electrode and antenna tip of ~ 1nm. The module that is an assembled array of rectennas with electrical connections is called a Monolithic Nanoscopic Tunnel Junction (MNTJ), which is shown in Fig. 12(c).

Two other antenna structures, the thin-wire terminated with a triangular tip (similar to Fig. 12) and a half-bowtie, are demonstrated in Figs. 13 and 14. Figs. 13 and 14 show the SEM images of a fabricated device module before ALD. The active sensor area (~400 x 200 μm) contains about 10,000 rectennas. The dimensions of the antennas of each of the modules were initially designed to optimize absorption in the red and IR. As mentioned, we fabricated these MNTJ arrays by conventional lithography and e-beam techniques. This process is then followed by selective ALD to yield junctions in the nanometer range. Such facilities are available in the Penn State Nanofabrication Facility at Penn State University Park.

Electro-optical characterization of these devices is ongoing both before and after ALD processing and will be reported in a subsequent publication (see Summary and Future Work). Preliminary images of the devices after selective ALD at the University of Connecticut show that the Cu successfully grew on the Pd tips and narrowed the gap. In Figure 15, we show SEM images of one of our modules with 10,000 rectennas, where the gap distance was narrowed from 70nm to about 20 nm. Note that the morphology of the point contact remains in the conformal process. Moreover, no growth was observed on the contact pad (Al) and surrounding SiO₂ areas, as expected.³⁷
**Fig. 13.** SEM image of the top view for the columnar antenna with triangular tip.

**Fig. 14.** SEM image of the top view for the triangular antenna.
(a) Before Cu ALD

(b) After Cu ALD

**Fig. 15.** (a) SEM image for the triangular antenna before ALD. The Tip-wall gap distance ~70nm; Tip length ~100nm; Wall-wall distance ~170nm. (b) SEM image at an intermediate stage of the ALD deposition. Tip-wall gap distance ~21nm; Tip length ~86.5nm; Wall-wall distance ~108nm. Energy-dispersive X-ray spectroscopy (EDX) data after Cu ALD show that the Cu successfully grew on the Pd tips and closed the gap. Also, no growth was observed on the contact pad (Al) and surrounding SiO2 areas, as expected. The inset is a magnified view of the tips showing the selective nature of the ALD deposition.

5. SUMMARY AND FUTURE WORK

We have surveyed developments related to the fabrication and theoretical understanding of nanoscale rectennas. The rectenna devices, based on the geometrically-asymmetric tunnel junction, can collect and rectify electromagnetic radiation, from the infrared through the visible regimes. Studies of electron transversal time and RC response time demonstrate that tunnel junctions formed with a sharp tip (early examples of which are the whisker diode and the STM probe) are capable of operating into the UV regime. Recent efforts to construct nanoscale antennas reveal a wealth of promising geometries and fabrication techniques (in thin-wire and patch antennas). Other recent experimental work confirms that nanorectennas are capable of not just receiving, but also rectifying, signals through the visible regime. A number of simulation studies by Mayer et al. not only demonstrate the viability of the geometrically-asymmetric tunnel junction, but also establish the importance of certain design parameters (choice of geometry and materials) that will be crucial in efforts to optimize such devices. In this concluding section, we present our own plans for expanding on the efforts already summarized. These plans include a process for fabricating individual nanoscale rectenna devices and also larger MNTJ arrays. The program is built around the use of standard lithography techniques and a novel use of atomic
layer deposition (ALD), and a rigorous program of IV characterization, computer simulation, and device optimization.

There is a need for a series of systematic experiments and theoretical work to understand more rigorously the interaction of electromagnetic radiation with nanoantennas and other nanostructures.\(^{38,39,40}\) At visible light frequencies the classical skin depth in metals is about 30 nm and metals become transparent in the UV, implying that classical theory is no longer applicable and quantum effects need to be considered. Electrical characterization of the MNTJs should be complemented by electron microscopy including SEM and high-resolution transmission electron microscopy (HRTEM). To understand scaling effects, devices should undergo electrical characterization including frequency response, I-V behavior, rectification ratio, and open-circuit voltage. In the following paragraphs, we briefly describe ongoing experimental and theoretical work using rectenna arrays with point-contact rectifying tunnel junctions. These studies will use a second round of refined MNJTJs currently being fabricated at the Penn State Nanofabrication Facility.

To demonstrate the spectral-behavior of the MNTJs, arrays will be irradiated by collimated laser light from the following single-wavelength sources: 475 nm, 532 nm, 642 nm, and 1064 nm. Once design features and device efficiency are determined and optimized, a solar simulator will be used to replicate broadband solar radiation. The light intensity of the solar simulator can be varied as can the wavelength, using a diffraction monochromator (0.5 nm resolution). Hence, the ability to control wavelength and intensity will provide a frequency-response spectrum for the device and allow the determination of practical limits of its rectifying behavior and efficiency.

I-V characteristics will be determined using lock-in amplifier techniques to isolate the photocurrent \(I_{\text{photo}}\) from the tunneling current under bias, as well as any thermally-induced contribution. To this end, the laser or solar simulator light can be modulated \((f \sim 200 \text{ Hz})\) with an optical chopper whose output frequency will be referenced by a lock-in amplifier. The light input to the device is then focused and directed to the MNTJ array. The rectenna junctions can be biased with a dc voltage plus a small ac component, with the dc swept over a range to produce both forward and reverse current. Thus, by using lock-in amplification one can simultaneously measure \(I_{\text{dc}}, \partial I / \partial V, \partial^2 I / \partial V^2\), and \(I_{\text{photo}}\) which is derived from the non-linear term in the current.\(^{13,25,31}\) The I-V characteristics will be determined under a variety of relevant conditions to fully benchmark the MNTJs: with and without incident light, as a function of polarization, angle of incidence, wavelength, and intensity. This data will determine the rectification ratio, \(I^+ / I^-\), and its dependence on the various rectenna parameters necessary to optimize the device structure.

Measurements of the MNTJs for open-circuit voltage, under the varying irradiation conditions mentioned above, will provide insight into the radiation-induced bias that initiates tunneling. We have performed preliminary open-circuit measurements on tungsten whiskers.\(^{21}\) Voltages of up to 100 mV are created with laser irradiation of \(\lambda = 1064 \text{ nm}\) and \(\lambda = 532 \text{ nm}\), in agreement with the results of Nguyen, \textit{et al.}\(^{7}\) This suggests that the electric fields are sufficient for producing tunneling currents on the order of nA per junction.

An additional outcome of I-V characterization is the determination of the external quantum efficiency \((\text{EQE} = P_{\text{out}} / P_{\text{in}})\) as a lower bound on the internal quantum efficiency \((\text{IQE} = P_{\text{out}} / P_{\text{absorbed}})\), which is critical to the evaluation of the rectenna’s potential as an energy harvesting device. This will be accomplished by splitting the incident light beam and sending half the light to a NIST-traceable calibration photodiode and the other half to the MNTJ. The photocurrent
responses of both beams will be measured simultaneously, which will help account for any intensity fluctuations or changes in the optical setup over time.\textsuperscript{41,42} Moreover, one can measure the incident and reflected radiation across the full spectrum to gauge the radiation actually absorbed by the MNTJ, thus providing an experimental value of the IQE.

In investigating device efficiency, it will be important to consider frequency, material, and geometry. For conventional photovoltaics, the DC power output is strongly bandgap and resistivity dependent. By contrast, the efficiency of the proposed rectenna devices depends primarily on the controllable geometry and the material parameters (e.g., electrical and optical constants) with no bandgap dependence for metallic emitters. Distinct advantages of the nanowire rectenna are the demonstrated fast response up to the green part of the spectrum, as well as the low resistivity, which reduces the heat losses in the system. For energy harvesting devices, these facts suggest an increased efficiency of the point-contact metallic nanowires over photovoltaic devices using standard MOM or Schottky diodes. The theoretical efficiency requires the determination of output DC power, which can, for example, be calculated using the model of Sullivan et al.\textsuperscript{17} and the quantum-mechanical formalism of Mayer et al.\textsuperscript{12}

One can provide an estimate of the efficiency of a nanowire rectenna irradiated with solar radiation. Output power is given by $P_{\text{out}} = J A_e V$, where $J$ is the field emitted current density, $A_e$ is the emitting area, and $V$ is the DC rectified voltage developed between the tip and base. The effective input power to the rectenna for the solar intensity is given by $P_{\text{in}} = I_s A_b$, where $I_s$ is the solar intensity of 0.1 W/cm\textsuperscript{2} and $A_b$ is the area of the beam intercepted by the nanowire. Therefore,

$$\eta = \frac{P_{\text{out}}}{P_{\text{in}}} = \frac{J A_e V}{(I_s A_b)} \quad (20)$$

To estimate these quantities, one can use the conventional Fowler-Nordheim equation

$$J = 1.54 \times 10^{-6} (F^2 / \varphi) \text{Exp}\left(-6.83 \times 10^7 \varphi^{3/2} / F\right), \quad (21)$$

where $F$ is the local field at the tip and $\varphi$ is the work function. The field values and the corresponding values of $V$ and $F$ are obtained from the work of Sullivan et al.\textsuperscript{14,17} The area of the emitting tip can be estimated from a standard model calculation for field emitters. The the solar irradiance, $I_s$, is equal to 0.1 W/cm\textsuperscript{2} and $A_b$ is approximately one-half the cylindrical surface area of the rectenna. For a tip with radius of 2 nm and field values consistent with Sullivan et al., ($\sim 3 \times 5 \times 10^7 V/cm$), the efficiency is $\eta \geq 0.50$.

This estimate suggests that efficiencies may equal or exceed those of competitive technologies. The calculation also suggests that the efficiency can be controlled by judicious choices of material (work function) and geometric parameters of the tip.

New simulations of these metal nanostructures are needed to determine their response and properties under electromagnetic illumination. While the behavior of individual rectennas of a particular morphology and gap spacing can be readily characterized by the methods already outlined, the ability to modify these parameters, fabricate new devices, and assess the scalability of MNTJ devices for the practical use as energy conversion devices is both cost- and time-prohibitive. Thus to better understand the response of individual junctions and the collective response of many rectenna junctions incorporated into MNTJ arrays, we will perform finite element modeling to simulate the response of the MNTJs to incident electromagnetic fields.
These computer simulations will use a commercially-available finite-element modeling (FEM) package such as COMSOL,\textsuperscript{43} and complement the work of Mayer, \textit{et al.},\textsuperscript{9,10,11,12,13} with the goal of providing design-improvement feedback. Recent work by researchers in the nanophotonics field have constructed useful nanoantenna models and derived simulation data important to the design of new experiments for proof of principle confirmation. For example, Zhou and co-workers, using COMSOL have investigated the plasmon-enhanced optical properties of metal nanoparticle-coated silicon nanowires for use as wired solar cells;\textsuperscript{44} similar work by Kildishev, \textit{et al.}, used an FEM model to establish the role of surface roughness in the optical response of plasmonic nanoantenna arrays.\textsuperscript{45} Data obtained from preliminary measurements of individual rectenna devices will be used as preliminary input for examining alternative geometries, junction electrode materials, gap spacing, and scaling over several orders of magnitude. The resulting simulation data will provide insight into the scattering and absorption of light by the MNTJ, as well as enable efforts to optimize device performance. The objective is that extrapolation of the results for MNTJ arrays whose aggregate size exceeds the wavelength of the incident radiation will accurately reflect the degree to which these devices can convert light into electricity on the macroscale.

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\[ \gamma_k = \omega \left( 2m/V_b e^2 \right)^{1/2} \]
where \( m \) is the electron mass, \( e \) the charge on the electron, \( E \) is the electric field strength generated by the laser and \( \omega \) is the angular frequency of the radiation. In the case of a metal, the height of the barrier, \( V_b \), is typically identified with the work function and for a semiconductor with the band gap. As would be expected a value of \( \gamma_k \gg 1 \) indicates multiphoton processes dominate (in the barrier and/or tunneling process), and, \( \gamma_k << 1 \) implies tunneling processes dominate.


In comparison, an estimate of the power density for a laser with a power output of 20mW and a beam diameter of about 300 μm is on the order of 20 W/cm².


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