

# Electron-emission materials: Advances, applications, and models

Daniele M. Trucchi and Nicholas A. Melosh, Guest Editors

Electron emission represents the key mechanism enabling the development of devices that have revolutionized modern science and technology. Today, science still relies on advanced electron-emission devices for imaging, electronics, sensing, and high-energy physics. New generations of emission devices are continuously being improved based on innovative materials and the introduction of novel physical concepts. Recent advances are highlighted by emerging low-work-function and low-dimensional materials with unusual electronic and thermal properties. Nanotubes, nanowires, graphene, and electron-emission models are discussed in this issue, as well as original mechanisms, such as the thermoelectronic effect, thermionic emission, and heat trap processes. Advances in electron-emission materials and physics are driving a renaissance in the field, both opening up new applications, such as energy conversion and ultrafast electronics, as well as improving traditional applications in electron imaging and high-energy science.

## Introduction

Electrons represent data carriers for computing and imaging as well as power carriers for electrical energy. As opposed to electron flow in solid-state materials, “free” electrons in a vacuum are unfettered by scattering and recombination mechanisms, providing advantages for electron transfer, manipulation, and imaging. Vacuum electron devices formed the core of early 20th century technology spanning from electronics to thermal-energy conversion, from microscopy to x-ray generation, from imaging to mass spectrometry, from linear accelerators to sensors. Stimulating, enhancing, and controlling emission of electrons and their subsequent flight in space and time has driven the development of low-work-function materials, high-aspect-ratio devices, nanostructures, and radiation-absorbing materials.

Electron emission has a rich history that played a pivotal role in vacuum-tube technology and has underpinned the development of modern electronic devices and circuits. At a fundamental level, electron emission relies on excitation of electrons above the material’s work function, typically 3–5 eV, allowing these energetic electrons to exit the material when they encounter the surface. Many different physical stimuli can supply this energy, including thermal-energy (coined as “thermion” emission by Thomas Edison in 1880), photons (the photoelectric effect made famous by Einstein),

ion or electron bombardment, and large electric fields, which, unlike the others, allow electron tunneling toward the vacuum (**Figure 1**). Understanding these processes led to the invention of the vacuum diode by J.A. Fleming in 1903,<sup>1</sup> and the Nobel Prize in Physics to O.W. Richardson in 1928 for the theoretical description and equation for thermionic emission.<sup>2</sup> Following on Fleming’s work, L. De Forest developed the vacuum tube “triode,” using a third terminal to provide current control and amplification,<sup>3</sup> leading to the subsequent electronic revolution.

Devices based on electron emission and emissive materials remain important objectives of 21st century science. Compared to early vacuum-tube technology, today’s applications require higher electron density, narrower electron-energy distribution, shorter emission times, and more efficient excitation. These requirements have propelled the development of new materials and physical emission mechanisms, often taking advantage of the unique electronic and thermal properties of low-dimensionality materials and nanoscale phenomena. The articles in this issue review a number of these new materials and applications.

## Recent advances in electron emission

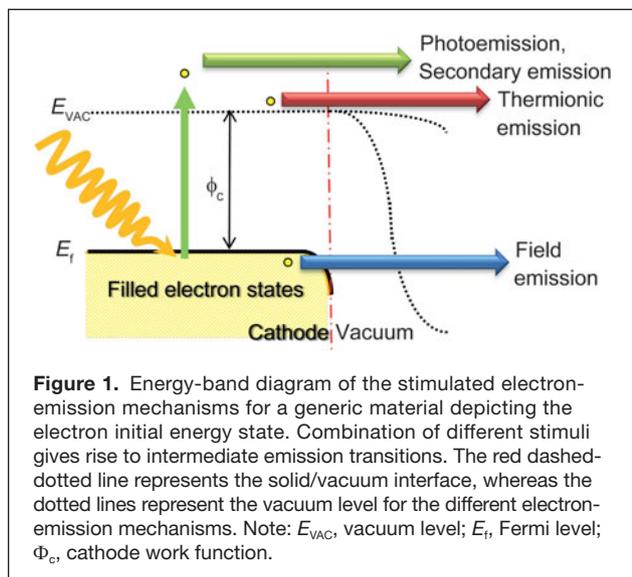
The different physical mechanisms inducing electron emission can be classified as photoelectron, secondary electron,

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thermionic, and field electron emission, or combinations of one or more of these. Each particular physical stimulus necessitates cathode materials with physical properties specifically engineered for optimal performance. This section will present an overview of the latest results on the development of advanced materials and related applications, categorized by the specific type of electron emission.

### Photoemission and secondary electron emission

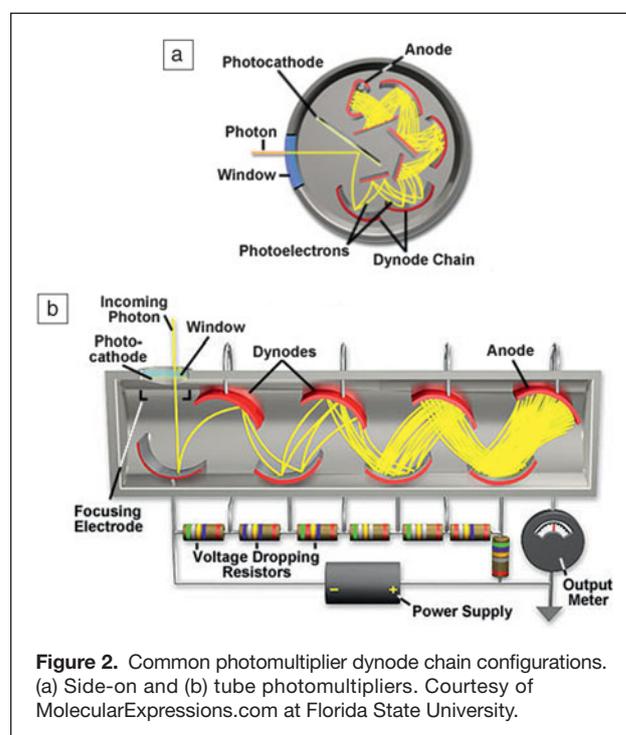
Photomultipliers remain the lowest noise, highest sensitivity, and fastest imaging systems. Used, for instance, in fluorescence and laser scanning confocal microscopy, they exploit electron emission from a photocathode and, successively, secondary emissions from electron multiplying stages. Photons to be detected induce the emission of electrons from a material, usually a thin layer in transmission mode. The emitted electron can be accelerated in a vacuum tube by an electric field toward an electron multiplier stage composed of a set of dynodes, which are basic components that emit several low-energy electrons (secondary electrons) when one high-energy electron (primary electron) impinges on their surfaces. **Figure 2** shows two common photomultiplier configurations. The sensitivity of photocathodes depends on the energy of impinging photons. Consequently, it is not possible to define the best possible photoemission material for a wide range of optical wavelengths.

III–V semiconductors with a bandgap engineered according to the corresponding photon energy band are the preferred solution for detecting visible and infrared (IR) radiation; these are also characterized by ultrafast response.<sup>4</sup> Cesium-coated GaAs operates at wavelengths up to 930 nm, whereas InGaAs extends the IR range up to 1700 nm. Ag–O–Cs materials are also used for visible–IR detection, with a preferred use in the near-IR region due to a higher sensitivity.<sup>5</sup> Multi-alkali-based technology covers the range from the near-IR to the ultraviolet (UV), whereas the Cs–Sb coatings operate from visible to UV with a lower quantum efficiency than the multi-alkali systems.

Conversely, Cs–Te technology achieves a high quantum efficiency, but in the limited 200–300 nm wavelength range. For UV wavelengths <200 nm, Cs–I technology is standard, whereas nano- and microstructured diamond are appealing candidates for future vacuum UV<sup>6</sup> and extended UV<sup>7</sup> photocathodes with hydrogen- or lithium-oxygen termination on the diamond surface,<sup>8</sup> owing to the high quantum efficiency induced by negative electron affinity.

These same concepts apply to electron multipliers in all-electron imaging systems (e.g., scanning electron microscopy).<sup>9</sup> Electron multipliers consist of several dynodes operating on secondary electron emission, where a number of low-energy electrons (secondary electrons) are emitted. Usually, the secondary electron emission occurs as a consequence of a primary beam of high-energy electrons, but it can include ions, alpha, and beta particles. Dynodes should have secondary electron-emission yield (the ratio between impinging and emitted electrons) as high as possible for the development of electron multiplier stages with high gain. The acceleration of electrons in each emission step is necessary to allow the primary beam to achieve a kinetic energy for which the emitting material shows the maximum of the secondary emission yield. Absolute yield value and primary electron energy for obtaining the maximum yield are the most important parameters.

A material with a high secondary electron-emission yield allows a reduction in the number of dynodes or a high total gain with the same number of dynodes. On the other hand, a material with a maximum yield occurring for low primary beam energy implies low applied voltages (i.e., safer operation) as well as low consumed power (i.e., high efficiency). Independent of geometry (reflection or transmission mode, with transmission



mode preferred owing to a minor degradation of the emitting surface due to the primary beam bombardment),<sup>10</sup> the optimal material shows low work function and large electron mean free path. Materials with a high secondary electron-emission yield are usually semiconductors or insulators, including alkali halides and alkaline earth compounds, or semiconductors with excellent transport properties such as Cs-O terminated silicon and GaAs, MgO, and Al<sub>2</sub>O<sub>3</sub>. While many of these coatings are simply evaporated onto the substrate, more controlled methods such as atomic layer deposition<sup>11</sup> and chemical vapor deposited materials such as hydrogen-terminated diamond<sup>12,13</sup> are promising.

A current ambitious scientific topic is the development of coatings with minimum secondary electron-emission yield for high-energy physics accelerator applications (e.g., Large Hadron Collider at Conseil Européen pour la Recherche Nucléaire or the Linac Coherent Light Source at Stanford Linear Accelerator Center). The aim is the suppression of undesired emissions caused by the primary beams impinging on the accelerator inner walls. In this case, low-cost large-area thin amorphous carbon coatings<sup>14</sup> as well as specific surface geometries<sup>15</sup> are under development.

### Thermionic emission

Thermionic emitters are used in a variety of applications, including high-frequency vacuum transistors for electronics, electron guns for scientific instrumentation, power electronics, x-ray generation, and energy converters from high-temperature sources and solar energy. Fundamentally, thermionic emission relies on heating the material to sufficient temperatures (usually >1200°C) such that a small number of electrons in the material have sufficient thermal energy to overcome the work function. Lowering the material's work function, such as with low-electron affinity coatings, allows emission at lower temperatures and is thus a critical component of thermionic devices. This is often accomplished by enclosing a Cs or Ba source that vaporizes at the operation temperature, coating the emitter within a vacuum chamber. This increases the size, complexity, and cost of the device. Thus, new concepts such as "heat" traps in carbon nanotube forests (see the Nojeh article in this issue) are interesting because they can rapidly heat to such high temperatures that work-function reduction is not necessary. Moreover, one- and two-dimensional (2D) conductors are enabling more efficient and smaller form factor devices that could replace solid-state devices for high-performance applications, as reviewed by Wei et al. in their article in this issue.

It was recognized early in the 20th century that thermionic emission from a hot surface could be an efficient way to convert high-temperature thermal energy directly into electricity,<sup>16</sup> with practical applications in the nuclear<sup>17</sup> and aerospace sectors.<sup>18,19</sup> One of the key barriers for widespread adoption of thermionic energy conversion has been its modest efficiency, ~10–15%. Theoretically, the efficiency could be quite high as the ideal Carnot efficiency of a heat engine operating at 1200°C is 82%, and there are no moving parts to cause friction. The limitation is

the space-charge effect—the electric field induced by the emitted electrons. In its simplest form, a thermionic convertor consists of two parallel plates, one of which (the cathode) is heated to thermionically emit electrons across a small vacuum gap to the other electrode (the anode). During the traversal across this vacuum gap, there is no positive charge to balance the electrons, generating a significant negative electrostatic charge that suppresses further emission. Overcoming this space-charge effect is a major objective for efficient energy-conversion devices. Several approaches are being pursued, including reducing the gap between the electrodes, positive compensation, and using neutral carriers. In this issue, Wanke et al. describe using modern microfabrication techniques to provide electrostatic neutralization, which could pave the way for highly efficient, compact, and scalable electricity generation.

Thermionic conversion was also recently revisited for concentrated solar-power applications. In the most direct incarnation, solar radiation is simply used as the heat source for the cathode, driven by the development of low-work-function emitters that can operate under moderate temperatures (~700°C),<sup>20</sup> and formulation of new concepts such as thermionic–photovoltaic<sup>21</sup> and thermionic–thermoelectric<sup>22,23</sup> combined hybrid devices. However, the availability of high-energy photons enables new combinations of photon and thermal mechanisms. A recent concept combined quantum photoexcitation together with thermionic emission, known as photon-enhanced thermionic emission (PETE).<sup>24</sup> In combination with a thermodynamic conversion stage, usually consisting of thermal engines capable of recovering and converting the waste heat, efficiencies close to 70% may be possible.<sup>25</sup> In addition, comparably high efficiencies can be achieved, unexpectedly, under isothermal conditions<sup>26</sup> (i.e., electrodes operating at the same temperature, which contradicts the basic principles of standard thermionic energy conversion that dictate a thermal gradient between the electrodes). Owing to the strong sensitivity to carrier lifetimes and absorption, III–V semiconductors are the preferred active materials for fabrication of PETE devices,<sup>27,28</sup> which have been investigated in terms of surface tailoring to more effectively capture sunlight<sup>29</sup> and in terms of thermally driven degradation of the emitting coating.<sup>30</sup> Interesting alternatives are low-work-function nanocrystalline,<sup>31</sup> polycrystalline,<sup>32</sup> and black diamond films,<sup>33,34</sup> based on surface nanotexturing to improve the optical and photoelectronic interaction of diamond with sunlight.<sup>35</sup>

### Field emission

Field electron emission, usually abbreviated to field emission, relies on the action of a strong electric field to reduce the energy barrier at the surface of the emitter. The height and width of the energy barrier for electron emission are reduced to such an extent that electrons begin tunneling from the surface. Since the electrons do not need elevated energy within the material (such as thermal excitation), field emitters are often defined as "cold" cathodes.

The performance of field emitters is generally evaluated based on three criteria—extraction field, brightness, and monochromaticity. The required electric field for extraction is influenced by the work function of the emitter as well as geometric field enhancement. The “turn-on” field exponentially decreases with the emitter-aspect ratio, given by the ratio of the height to tip radius. Tall, sharp tips are preferable, making nanomaterials ideal candidates. Small emitter area also leads to higher brightness, measured as the emitted current per unit of angle of divergence, applied voltage, and emitting area. Just as important is the energy distribution of the electrons, known as the monochromaticity. Imaging in scanning and transmission electron microscopes is particularly sensitive to “chromatic aberrations” in the energy distribution, thus a narrow kinetic energy spread is desirable.

New nanofabrication tools emerging from the semiconductor industry are used to create nanoscale field emitters with components separated by nanometer vacuum gaps. Examples of vacuum silicon nanodevices are gate-insulated channel transistors with a cutoff frequency of 0.46 THz,<sup>36</sup> a value comparable only to graphene-based transistors and almost an order of magnitude higher than that of GaN technology, and vacuum-channel transistors with emission current densities as high as  $10^5$  A/cm<sup>2</sup> originating from 2D electron gases.<sup>37</sup> **Figure 3** shows vertical and planar emitter configurations, developed analogously to the metal oxide semiconductor field-effect transistor technology. Nanostructures engineered to allow surface plasmon resonances can be used to electro-optically emit electrons for semiconductorless high-speed micro- and

optoelectronic devices.<sup>38</sup> The capability to develop small form factor nanostructures coupled with low-work-function materials may begin to level the playing field between vacuum and solid-state devices. This is still an open scientific question.

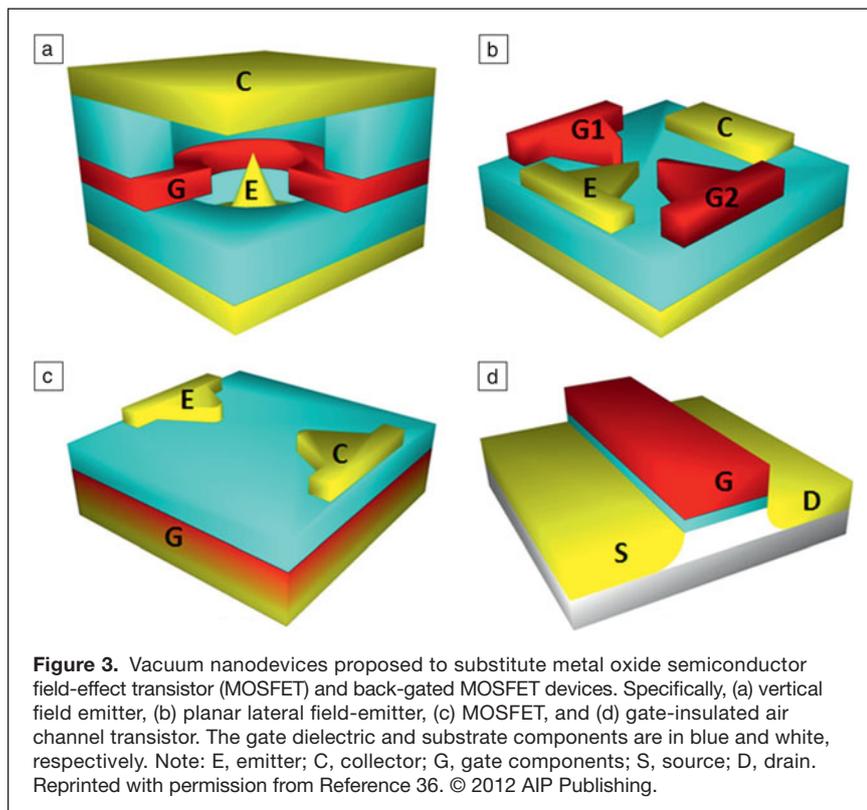
One of the first demonstrations of the benefits of nanotechnology was in low-voltage field emission from carbon nanotubes<sup>39,40</sup> due to the high field enhancement of the nanometer-scale tips. LaB<sub>6</sub> nanowires<sup>41</sup> and other nanomaterials<sup>42</sup> are also being extensively investigated as cold cathodes. However, the large geometric field enhancement conversely leads to extreme sensitivity to slight changes in shape, even at the nanometer level. Stable emission is thus difficult to achieve as local heating, atomic desorption, and electromigration at the tip can dramatically change emission characteristics. Today, field emission from nanostructured materials is still a strong materials science research focus. Zhang et al. review the latest developments in their article in this issue.

### Emerging materials and applications

The strong interest of the scientific and technological communities in the development of low-dimensional materials is evident in the increasing and accelerating number of electron-emission publications in recent years. As seen from the collection of articles in this issue, the dominant theme has been nanomaterials and nanoscale fabrication. Thermionic and field emission are experiencing a transition toward 2D materials, especially carbon-based materials. Graphene,<sup>43</sup> discussed by Ang et al. in their article in this issue as a tunable electron emitter for vacuum nanoelectronics, represents a clear example

of this transition. Owing to the reduced dimensionality, new fundamental models describing and predicting thermionic and field emission are necessary, which readers will find discussed in the articles in this issue. Apart from carbon-based materials, the longer excited lifetimes and potentially low work function of perovskites with controlled surface termination<sup>44</sup> are promising future directions.

Another emerging application for photocathodes is the direct exploitation of solar radiation, for example, photocathodes and photoanodes for water splitting aimed at hydrogen production induced by sunlight. In this case, electrons are not emitted into the vacuum, but into the aqueous electrolyte solution of a photoelectrochemical cell, engineered as a device able to suitably produce a sufficient voltage to split water molecules.<sup>45</sup> Significant research is being performed in this field for developing not only an effective approach for the collection of sunlight and efficient electron emission, but also improved corrosion resistance, carrier mobility, and stability of the electrode surfaces. Recent advances suggest semiconductor,<sup>46</sup> boron-doped diamond foam,<sup>47</sup> and organic–inorganic



electrodes,<sup>48</sup> possibly operating at high temperatures, may be feasible for solar radiation systems.<sup>49,50</sup>

## Conclusion

This issue of *MRS Bulletin* is inspired by the concept that new materials and physics can reinvigorate the field of electron emission. The exciting possibility to exploit 2D materials for electron emission creates new scenarios, which, in combination with different emission mechanisms, allows advanced applications, some of which improve upon classical devices, and some that may not have been invented yet.

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