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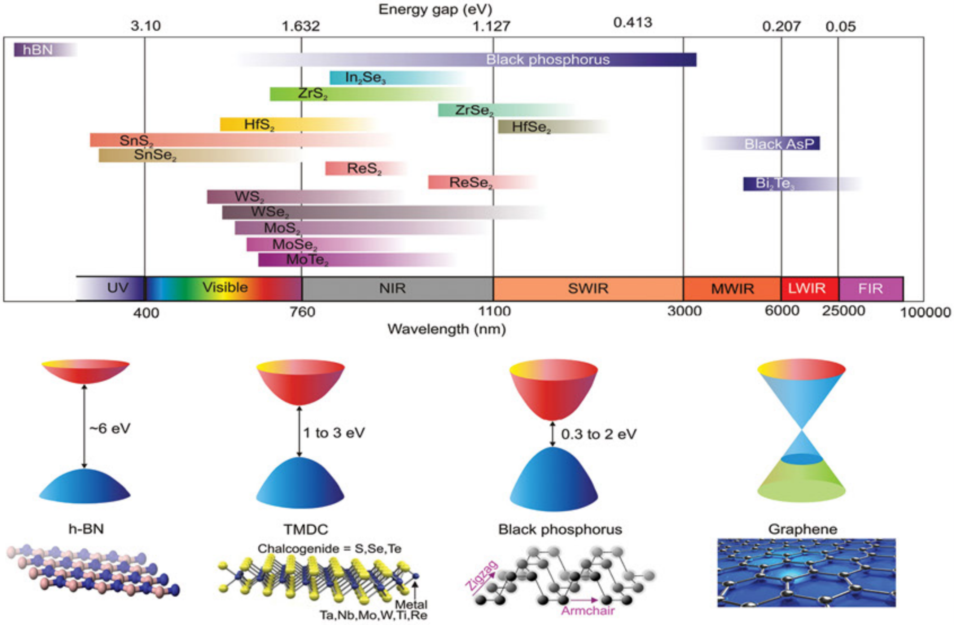

2D MATERIALS FOR INFRARED AND TERAHERTZ DETECTORS

Antoni Rogalski

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2D Materials for Infrared and Terahertz Detectors

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Contents

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Preface

Since the discovery of graphene, its applications to electronic and optoelectronic devices have been intensively and thoroughly researched. Its extraordinary and unusual electronic and optical properties allow graphene and other two-dimensional (2D) materials to be promising candidates for infrared (IR) and terahertz (THz) photodetectors. Until now, however, their place in the wide-infrared detector family has not been evaluated and this topic is generally omitted from the review literature.

The main goal of this book is to provide a critical view on the present state of 2D-material photodetector technologies and on future developments with respect to global competition with existing industrially mature material detector systems, such as HgCdTe, InGaAs, type-II superlattice III-V compounds, and microbolometers. This book also considers the challenges facing development of focal plane arrays for the future. Special attention is paid toward the main trends in development of arrays in the near future, such as increases in pixel count to above 10^8 pixels, with pixel size decreasing to about 5-µm, mostly for uncooled infrared arrays. Until now, these questions have not been considered in literature reviews devoted to 2D-material IR and THz detectors.

Most of the 2D layered semiconducting material photodetectors operate at the visible and near-infrared regions. However, the thrust of this book is mainly directed to effective IR and THz detectors, based on 2D materials. This book

- gives brief accounts of the different types of 2D materials used in the fabrication of IR and THz detectors,
- describes advantages and disadvantages of 2D materials as IR and THz detectors,

- sets in order the performance of 2D material IR and THz detectors among the family of common commercially available detectors,
- tries to predict the future role for 2D materials in the family of detectors,
- predicts the main trends in development of arrays in the near future.

The number of published papers devoted to the use of 2D materials as sensors is huge. However, the authors of these papers mainly address their work to researchers involved in investigations of 2D materials. In this book, the position of 2D material detectors is considered in comparison with the present state of conventional infrared and terahertz detectors offered on the global market. In this way, the book gives an overview of the performance of emerging 2D material detectors, comparing them with traditionally and commercially available ones under different conditions, including high operating-temperature conditions.

This monograph is divided into eight chapters. After the introduction, two chapters (2 and 3) describe detector characterization and fundamentals of detection mechanisms for both thermal and photon detectors, including detector performance limits. These initial chapters provide a tutorial introduction to the technical topics that are necessary for a thorough understanding of the different types of detectors and systems. In Chapter 3, a new reference benchmark, the so-called "Rule 19", is introduced for prediction of the performance of background-limited HgCdTe photodiodes, operated near room temperature. This rule is subsequently addressed in the following chapters (6, 7, and 8) as a benchmark against which to compare alternative 2D material technologies.

In Chapter 4, topics are considered which are almost completely omitted by the scientific community researching 2D detector materials, including future trends in the development of focal plane arrays. Taking into account the early stages of development and manufacturability, such considerations are essential to make a realistic assessment of the prospects for subsequent commercialization of 2D-material photodetectors.

The next four chapters (5, 6, 7, and 8) briefly describe the fundamental properties of graphene-based materials and other 2D materials, and the performance parameters (such as responsivity, detectivity, and response time) of detectors fabricated with these materials, in comparisons between 2D material-based detectors and traditional detectors on the global market, including both experimental data and theoretical predictions. Final conclusions predict the likely place of 2D material-based detectors in the wide-IR detector family, in the near future.

The presentation level of this book is suitable for graduate students in physics and engineering, who have received background training in modern solid-state physics and electronic circuits. This book would also be of interest to individuals working with aerospace sensors and systems, remote sensing, thermal imaging, military imaging, optical telecommunications, infrared spectroscopy, and light detection and ranging

This book, I hope, will provide a timely and appropriate analysis of the latest developments in 2D- material infrared and THz detector technology and a basic insight into the fundamental processes important to evolving detection techniques. The book covers different types of detectors, including the relevant aspects of theory, types of materials, their physical properties, and detector fabrication.

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CHAPTER 1

Introduction

Infrared (IR) radiation itself was unknown until 220 years ago, when Herschel's experiment with the thermometer was first reported. The first detector consisted of a liquid in a glass thermometer with a specially blackened bulb, to absorb radiation. Herschel built a crude monochromator that used a thermometer as a detector, so that he could measure the distribution of energy in sunlight [1].

The early history of IR was reviewed about 60 years ago in two well-known monographs [2,3]. Much historical information can be also found in more recently published papers [4,5]. The initial infrared detectors were based on the class of thermal detectors: thermometers, thermocouples, and bolometers [6]. In 1821, T.J. Seebeck discovered the thermoelectric effect, and soon afterward, in 1829, L. Nobili created the first thermocouple. In 1833, M. Melloni modified the thermocouple and used bismuth and antimony for its design [7]. Then, in 1835, Nobili, together with Melloni, constructed a thermopile capable of sensing a person 10 m away. The third type of thermal detector, the bolometer/thermistor, was invented by S.P. Langley in 1878. By 1900, his bolometer was 400 times more sensitive than his first efforts, and his latest bolometer could detect the heat from a cow at a distance of ¼ mile [8].

The photoconductive effect was discovered by W. Smith in 1873, when he experimented with selenium as an insulator for submarine cables [9]. This discovery provided a fertile field of investigation for several decades, though most of the effort was of doubtful quality. By 1927, over 1500 articles and 100 patents had been published on photosensitive selenium [10]. Work on the IR photovoltaic effect in naturally occurring lead sulfide, or galena, was first published by Bose in 1904 [11]; however, the IR photovoltaic effect was not exploited in a radiation detector for several more decades.

The photon detectors were developed in the twentieth century. The first IR photoconductor was developed by T.W. Case in 1917 [12]. He discovered that a substance composed of thallium and sulfur exhibited photoconductivity. Later, he found that the addition of oxygen greatly enhanced the response [13]. However, the instability of the resistance in the presence of light or polarizing voltage, the loss of responsivity due to overexposure to light or high noise, its sluggish response, and the lack of reproducibility seemed to be inherent weaknesses.

Since about 1930, the development of IR technology has been dominated by photon detectors. In about 1930, the appearance of the Cs-O-Ag phototube, with more stable characteristics, discouraged further development of photoconductive cells to a great extent until about 1940. At that time, interest in improved detectors had begun [14,15]. In 1933, Kutzscher, at the University of Berlin, discovered that lead sulfide (from natural galena found in Sardinia) was photoconductive and had a response to about 3 μm. This work was, of course, carried out under great secrecy and the results were not generally known until after 1945. Lead sulfide was the first practical IR detector deployed in a variety of applications during the war. In 1941, Cashman improved the technology of thallous sulfide detectors, which led to successful production [16]. After success with thallous sulfide detectors, Cashman concentrated his efforts on lead sulfide and, after World War II, found that other semiconductors of the lead salt family (PbSe and PbTe) showed promise as IR detectors [17]. Lead sulfide photoconductors were brought to the manufacturing stage of development in Germany in about 1943. They were first produced in the United States at Northwestern University, Evanston, Illinois in 1944 and, in 1945, at the Admiralty Research Laboratory in England [17].

Many materials have been investigated in the IR field. Observing a history of the development of the IR detector technology, a simple theorem, after Norton [18], can be stated: “All physical phenomena in the range of about 0.1–1 eV can be proposed for IR detectors.” Among these effects are: thermoelectric power (thermocouples), change in electrical conductivity (bolometers), gas expansion (the Golay cell), pyroelectricity (pyroelectric detectors), photon drag, the Josephson effect (Josephson junctions, SQUIDs), internal emission (PtSi Schottky barriers), fundamental absorption (intrinsic photodetectors), impurity absorption (extrinsic photodetectors), low-dimensional solids [superlattice (SL), quantum well (QW), and quantum dot (QD) detectors], different types of phase transitions, and so on.

Figure 1.1 gives approximate dates for significant developments for the materials mentioned. The years during World War II saw the origins of modern IR detector technology, supported by the discovery of the transistor in 1947 by W. Shockley, J. Bardeen, and W. Brattain [19]. Recent success in applying IR technology to remote sensing problems has been made possible by the successful development of high-performance IR detectors over the past seven decades. Photon IR technology, combined with semiconductor material science, photolithography technology developed for integrated circuits, and the impetus of Cold War military preparedness, propelled extraordinary advances in IR capabilities within a short period of time during the past century [20].

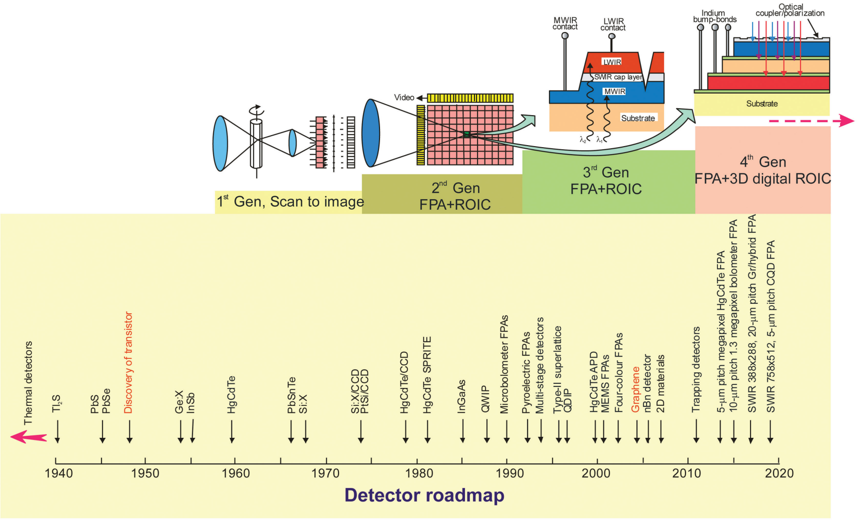

FIGURE 1.1 The history of the development of IR detectors and systems. For principal military and civilian applications, four generation systems can be considered: first-generation (scanning systems), second-generation (staring systems, electronically scanned), third-generation (staring systems, with large number of pixels and two-color functionality), and fourth-generation (staring systems with very large number of pixels, multi-color functionality, 3D ROIC, and other on-chip functions) systems, offering other functions, e.g. better radiation/pixel coupling, avalanche multiplication in pixels, and polarization/phase sensitivity.

1.1 HISTORICAL ASPECTS OF MODERN INFRARED TECHNOLOGY

During the 1950s, IR detectors were built using single-element-cooled lead salt detectors, primarily for anti-air missile seekers. Usually, lead salt detectors were polycrystalline and were produced by vacuum evaporation and chemical deposition from a solution, followed by a post-growth sensitization process [17]. The preparation process of lead salt photoconductive detectors was usually not well understood, and reproducibility could be achieved only after following well-tried recipes. The first extrinsic photoconductive detectors were reported in the early 1950s [21], after the discovery of the transistor, which stimulated a considerable improvement in the growth and material purification techniques. Since the techniques for controlled introduction of impurities became available for germanium earlier, the first high-performance extrinsic detectors were based on germanium. Extrinsic, photoconductive response from copper, zinc, and gold impurity levels in germanium gave rise to devices using the 8- to 14-μm longwave IR (LWIR) spectral window and beyond to the 14- to 30-μm very longwave IR (VLWIR) region. The extrinsic photoconductors were widely used at wavelengths beyond 10 μm, prior to the development of the intrinsic detectors. They must be operated at lower temperatures to achieve performance similar to that of intrinsic detectors, and a sacrifice in quantum efficiency is required to avoid thick detectors.

In 1967, the first comprehensive extrinsic Si detector-oriented paper was published, by Soref [22]. However, the state of the extrinsic Si was not changed significantly. Although Si has several advantages over Ge (namely, a lower dielectric constant, giving shorter dielectric relaxation times and lower capacitance, higher dopant solubility, a larger photoionization cross section for greater quantum efficiency, and a lower refractive index for lower reflectance), these were not sufficient to warrant the necessary development efforts needed to bring it to the level of the by-then highly developed Ge detectors. After the concept lay dormant for about 10 years, extrinsic Si was reconsidered after the invention of charge-coupled devices (CCDs) by Boyle and Smith [23]. In 1973, Shepherd and Yang [24] proposed the metal-silicide/silicon Schottky barrier detectors. For the first time, it became possible to have much more sophisticated readout schemes, so that both detection and readout could be implemented in one common silicon chip.

At the same time, rapid advances were being made in narrow-bandgap semiconductors, that would later prove useful in extending wavelength capabilities and improving sensitivity. The first such material was InSb, a member of the newly discovered III-V compound semiconductor family. The interest in InSb stemmed, not only from its small energy gap, but also from the fact that it could be prepared in single crystal form, using a conventional technique. The end of the 1950s and the beginning of the 1960s saw the introduction of narrow-gap semiconductor alloys in III–V ($InAs_{1-x}Sb_x$), IV–VI ($Pb_{1-x}Sn_xTe$), and II–VI ($Hg_{1-x}Cd_xTe$) material systems. These alloys allowed the bandgap of the semiconductor, and hence the spectral response of the detector, to be custom tailored for specific applications. In 1959, research by Lawson and coworkers [25] triggered the development of variable-bandgap $Hg_{1-x}Cd_xTe$ (HgCdTe) alloys, providing an unprecedented degree of freedom in IR detector design. This first paper [25] reported both photoconductive and photovoltaic response, extending out to 12 μm in wavelength. Soon thereafter, working under a U.S. Air Force contract with the objective of devising an 8–12 μm background-limited semiconductor IR detector that would operate at temperatures as high as 77 K, the group, led by Kruse, at the Honeywell Corporate Research Center in Hopkins, MN, developed a modified Bridgman crystal growth technique for HgCdTe. They soon reported both photoconductive and photovoltaic detection in rudimentary HgCdTe devices [26].

The fundamental properties of narrow-bandgap semiconductors (high optical absorption coefficient, high electron mobility, and low thermal generation rate), together with the capability for bandgap engineering, made these alloy systems almost ideal for a wide range of IR detectors. The difficulties in growing HgCdTe material, due significantly to the high vapor pressure of Hg, encouraged the development of alternative detector technologies over the past 40 years. One of these was PbSnTe, which was vigorously pursued in parallel with HgCdTe in the late 1960s and early 1970s [27–29]. PbSnTe was comparatively easy to grow, and high-quality LWIR photodiodes were readily demonstrated. However, in the late 1970s, two factors led to the abandonment of PbSnTe detector work: high dielectric constants and large mismatch of coefficient of thermal expansion (CTE) with Si. Scanned IR imaging systems of the 1970s required relatively fast response times, to avoid smearing the scanned image in the scan direction. With the trend today toward staring arrays, this consideration might be less important than it was when first-generation systems were being designed. The second drawback, a large CTE, can lead to failure of the indium bonds in hybrid structures (between the silicon readout and the detector array) after repeated thermal cycling, from room temperature to the cryogenic temperature of operation.

The material technology development was and continues to be primarily for military applications. In 1956, Texas Instruments had begun research on IR technology, which led to the signing of several contracts for a linear scanner, and subsequently to the invention of the first forward-looking infrared (FLIR) camera in 1963. As photolithography became available in the early 1960s, it was used to make IR detector arrays. Linear array technology was first applied to PbS, PbSe, and InSb detectors. The discovery in the early 1960s of extrinsic Hg-doped germanium [30] led to the first FLIR systems operating in the LWIR spectral window, using linear arrays. Because the detection mechanism was based on an extrinsic excitation, it required a two-stage cooler to operate at 25 K. The cooling requirements of intrinsic narrow- bandgap semiconductor detectors are much less stringent. Typically, to obtain the background-limited performance (BLIP), detectors for the 3–5 μm spectral region are operated at 200 K or less, while those for the 8–14 μm region are operated at the temperature of liquid nitrogen. In the late 1960s and early 1970s, the first-generation linear arrays of intrinsic HgCdTe photoconductive detectors were developed, in which an electrical contact for each element of a multielement array is brought off the cryogenically cooled focal plane to the outside, where there is one electronic channel at ambient temperature for each detector element Fig. 1.1). In 1972, Texas Instruments invented the HgCdTe Common Module concept, which contributed to a significant cost reduction and allowed for the reuse of common components. These allowed LWIR FLIR systems to operate with a single-stage cryoengine, making the systems much more compact and lighter, and consuming significantly less power.

Early assessment of the concept of the second-generation system showed that PtSi Schottky barriers, InSb, and HgCdTe photodiodes or high-impedance photoconductors, such as PbSe and PbS, and extrinsic silicon detectors were promising candidates because they had impedances suitable for interfacing with the field-effect transistor (FET) input of readout multiplexes. Photoconductive HgCdTe detectors were not suitable due to their low impedance and high-power dissipation on the focal plane. A novel British invention, the SPRITE detector [31,32], extended conventional photoconductive HgCdTe detector technology by incorporating signal time delay and integration (TDI) within a single elongated detector element. Such a detector replaces a whole row of discrete elements of a conventional serial-scanned detector, external associated amplifiers, and time-delay circuitry. Although only used in small arrays of about 10 elements, these devices have been produced in the thousands.

In the late 1970s and through the 1980s, HgCdTe technology efforts focused almost exclusively on photovoltaic device development, because of the need for low power dissipation and high impedance in large arrays to interface with readout input circuits. The emergence of advanced epitaxial techniques [molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD)], combined with the photolithography process, revolutionized the IR detector system industry, enabling the design and fabrication of complex focal plane arrays (FPAs). These efforts are finally paying off, with the birth of HgCdTe second-generation IR systems, that provide large two-dimensional (2D) arrays in both linear formats, with time delay and integration (TDI) for scanning imagers, and in square and rectangular formats for staring arrays. At the present stage of development, staring arrays have about 10^8 elements and are scanned electronically by circuits integrated with the arrays. It is predicted that larger focal planes will be possible, constrained by budgets rather than by technology [33]. These 2D arrays of photodiodes, connected with indium bumps to a readout integrated circuit (ROIC) chip as a hybrid structure, are often called a sensor chip assembly (SCA).

The first megapixel hybrid HgCdTe FPAs were fabricated in the mid-1990s. However, present HgCdTe FPAs are limited by the yield of arrays, which increases their cost. In such a situation, alternative alloy systems for infrared detectors, such as quantum well infrared photodetectors (QWIPs) and type-II superlattices (T2SLs), are being evaluated.

Recently, considerable progress has been made toward III-V antimonide-based low-dimensional solid development and device design innovations. Their development results from two primary motivations: the perceived challenges of reproducibly fabricating high-operability HgCdTe FPAs at reasonable cost, and theoretical predictions of lower Auger recombination for T2SL detectors, compared with HgCdTe. Lower Auger recombination translates into a fundamental advantage for T2SL over HgCdTe in terms of lower dark current and/or higher operating temperatures, provided that other parameters, such as Shockley-Read-Hall lifetimes, are equal. Recently, Raytheon's III-V T2SL (type II superlattice)/nBn detectors have also reached a level of maturity that enabled the company to win the contract for the next-generation Distributed Aperture System (DAS) for the F-35 Joint Strike Fighter [34].

HgCdTe has inspired the development of the three “generations” of detector devices. Third-generation devices are defined here to encompass the more exotic device structures, embodied in two-color detectors and hyperspectral arrays, which are now in production programs. For example, Raytheon fabricates the eLRAS3 system (Long Range Scout Surveillance System), which provides the real-time ability to detect, recognize, identify, and geo-locate distant targets outside the zone of threat acquisition. This high-definition high-resolution FLIR (also called 3rd Gen FLIR) combines HgCdTe longwave and mid-wave infrared arrays.

The first three generations of imaging device systems rely primarily on planar FPAs. We are currently dealing with the fourth-generation staring systems, in which the main features are to be high resolution (with a very large number of pixels, above 10^8), multi-color functionality, three-dimensional readout integration circuits (3D ROIC), and other integrated functions, e.g., better radiation/pixel coupling, avalanche multiplication in pixels, and polarization/phase sensitivity. The evolution of the fourth generation is inspired by the most famous visual systems, which are biological eyes. A solution, based on the Petzval-matched curvature, allowing the reduction of field curvature aberration, e.g. bonding the detectors to flexible or curved molds, has been proposed [35]. In addition, such a system combines such advantages as a simplified lens system, electronic eye systems, and wide field-of-view [36,37]. The colloidal quantum dot (CQD) [38] and 2D layered material photodetectors [39], fabricated on flexible substrates, are promising materials with which to overcome technical challenges in the development of fourth-generation IR systems. The unique and distinctive optoelectronic properties of graphene and related two-dimensional (2D) materials create a new platform for a variety of photonic applications, including infrared and terahertz photodetectors. In particular, there is growing interest in 2D materials for sensors, that have the potential to operate at room temperature.

As was mentioned previously, the development of IR technology has been dominated by photon detectors since about 1930. However, photon detectors require cryogenic cooling. This is necessary to prevent the heat generation by the charge carriers. The thermal transitions compete with the optical ones, making uncooled devices very noisy. The cooled thermal camera usually uses the Stirling cycle cooler, which is the most expensive component of the photon detector IR camera. Cooling requirements are the main obstacle to the widespread use of IR systems based on semiconductor photon detectors, making them bulky, heavy, expensive, and inconvenient to use.

The use of thermal detectors for IR imaging has been the subject of research and development for many decades. However, in comparison with photon detectors, thermal detectors have been considerably less exploited in commercial and military systems. The reason for this disparity is that thermal detectors are popularly believed to be rather slow and less sensitive in comparison with photon detectors. As a result, the worldwide effort to develop thermal detectors has been extremely small, relative to that of the photon detectors.

It must not be inferred from the preceding outline that work on thermal detectors has not been actively pursued. Indeed, some interesting and important developments have taken place along this line. In 1947, for example, Golay constructed an improved pneumatic infrared detector [40]. This gas thermometer has been used in spectrometers. The thermistor bolometer, originally developed by Bell Telephone Laboratories, has found widespread use in detecting radiation from low-temperature sources [41,42]. The superconducting effect has been used to make extremely sensitive bolometers.

Thermal detectors have also been used for infrared imaging. Evaporographs and absorption-edge image converters were among the first non-scanned IR imagers. Originally, an evaporograph was employed in which the radiation was focused onto a blackened membrane coated with a thin film of oil [43]. The differential rate of evaporation of the oil was proportional to the radiation intensity. The film was then illuminated with visible light to produce an interference pattern corresponding to the thermal picture. The second thermal-imaging device was the absorption-edge image converter [44]. Operation of this device was based upon utilizing the temperature dependence of the absorption edge of the semiconductor. The performance of both imaging devices was poor because of the very long time constraint and the poor spatial resolution. Despite numerous research initiatives and the attractions of ambient-temperature operation and low cost-potential, thermal detector technology has enjoyed limited success, in competition with cooled photon detectors, with respect to thermal imaging applications. A notable exception was the pyroelectric vidicon (PEV) [45], which was widely used by firefighting and emergency service organizations. The PEV tube can be considered analogous to the visible television camera tube, except that the photoconductive target is replaced by a pyroelectric detector and germanium faceplate. Compact, rugged PEV imagers have been offered for military applications but suffer the disadvantage of short tube-life and fragility, particularly the reticulated vidicon tubes, which are required for enhanced spatial resolution. The advent of the staring FPAs, however, marked the development of devices that would someday make uncooled systems practical for many, especially commercial, applications. The defining effort in this field was undertaken by Texas Instruments with contractual support from the Army Night Vision Laboratory [5]. The goal of this program was to build a staring FPA system based on ferroelectric detectors of barium strontium titanate. Throughout the 1980s and early 1990s, many other companies developed spatial devices based on various thermal detection principles.

The second revolution in thermal imaging began at the end of the twentieth century. The development of uncooled IR arrays, capable of imaging scenes at room temperature, has been an outstanding technical achievement. Much of the technology was developed under classified military contracts in the United States, so the public release of this information in 1992 surprised many in the worldwide IR community [46]. There has been an implicit assumption that only cryogenic photon detectors, operating in the 8–12 μm atmospheric window, had the necessary sensitivity to image objects at room temperature. Although thermal detectors have been little used in scanned imagers, because of their slow response, they are currently of considerable interest for 2D electronically addressed arrays, where the bandwidth is low and where the ability of thermal devices to integrate over a frame time is an advantage [47– 52]. Much recent research has focused on both hybrid and monolithic uncooled arrays and has yielded significant improvements in the detectivity of both bolometric and pyroelectric detector arrays. Honeywell has licensed bolometer technology to several companies for the development and production of uncooled FPAs for commercial and military systems. At present, compact megapixel microbolometer cameras are produced by Raytheon, L-3 Communications, FLIR, and DRS in the United States. The U.S. government allows these manufacturers to sell their devices to foreign countries, but not to divulge manufacturing technologies. Later on, several countries, including the United Kingdom, France, Japan, Israel, Korea, and China have “picked up the ball”, determined to develop their own uncooled imaging systems. As a result, although the United States has a significant lead, some of the most exciting and promising developments for low-cost uncooled IR systems in the future may come from non-U.S. companies (e.g., microbolometer FPAs with series p-n junctions, developed by Mitsubishi Electric).

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Colloidal Quantum Dot Infrared Detectors

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