

Plasmonic Organic Photovoltaic Devices on Transparent Carbon Nanotube Films

Emmanuel Kymakis, *Member, IEEE*, Emmanuel Stratakis, Emmanuel Koudoumas, and Costas Fotakis

Abstract—We have explored the effect of Au-nanoparticle (NP)-induced surface plasmons on the performance of polymer–fullerene photovoltaic devices utilizing single-walled carbon-nanotube (SWNT) films as the transparent electrode for hole collection. The Au NPs were produced by ultrafast laser ablation in liquids and were incorporated on the devices in a SWNT/poly(3,4-ethylene-dioxythiophene):poly(4-styrenesulfonate)/NP/poly(3-hexylthiophene):[6,6]-phenyl-C61-butyric acid methyl ester/Al configuration. The NP incorporation leads to a power conversion efficiency improvement of 70%. This increase can be attributed to the improved photocurrent and fill factor due to an enhanced exciton generation rate of the photoactive layer caused by localized surface-plasmon resonances of the conduction electrons within the NPs. This argument was supported by the combinatorial study of the optical properties of the NPs and the photon-to-electron conversion efficiency of the devices.

Index Terms—Nanocomposites, nanotubes, photovoltaic cells, plasmonic.

I. INTRODUCTION

ORGANIC photovoltaic devices have the potential to revolutionize the production of photovoltaic devices by offering a low-cost, environmental-friendly, and versatile alternative to their silicon counterparts, which currently dominate the world market. Their advantage lies particularly in the area of small-scale power sources, such as applications in mobile devices and wearable electronics, in which lightweight, flexible, and mechanically robust materials are needed [1]. To date, the most efficient and widely studied donor/acceptor system is the poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) [2], [3]. An interpenetrating network of nanoscale domains is formed, in which the photogenerated excitons are dissociated at the polymer/fullerene interfaces, and the electrons hop across to the PCBM to the negative electrode, while the holes travel through the polymer to the positive electrode.

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E. Kymakis and E. Koudoumas are with the Department of Electrical Engineering and the Center of Materials Technology and Photonics, Technological Educational Institute of Crete, 710 04 Crete, Greece (e-mail: kymakis@staff.teicrete.gr).

E. Stratakis is with the Institute of Electronic Structure and Laser, Foundation for Research and Technology–Hellas, 711 10 Heraklion, Greece, and also with the Department of Materials Science and Technology, University of Crete, 710 03 Heraklion, Greece.

C. Fotakis is with the Institute of Electronic Structure and Laser, Foundation for Research and Technology–Hellas, 711 10 Heraklion, Greece, and also with the Department of Physics, University of Crete, 710 03 Heraklion, Greece.

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A drawback of this configuration is the low optical absorption of the photoactive P3HT:PCBM layer in the near infrared (IR), which leads to a poor spectral mismatch with the terrestrial solar spectrum. An interesting approach for tackling this problem is the incorporation of metal nanoparticles (NPs) in order to increase the absorption of the device due to the high electromagnetic-field strength in the vicinity of the excited surface plasmons [4].

Surface plasmons are electromagnetic surface waves that can propagate at the interface between a metal and a dielectric material. The excitation of surface plasmons by light is denoted as a localized surface-plasmon resonance (LSPR) for nanometer metallic NPs, sized smaller than the incident-light wavelength. The strong optical extinction of the LSPR emerges in the visible or near-IR spectral regions due to driven collective electron oscillations, causing a local enhancement in the electromagnetic field. Hence, the incorporation of such NPs has the potential to significantly enhance the light absorption of the photoactive layer. This argument has been recently validated by several groups [5]–[7].

Recently, several groups have come with the idea of using carbon-nanotube (CNT) thin films as the transparent conductive electrode replacing the conventional indium–tin oxide (ITO) electrode in organic photovoltaic devices [8]–[11]. Highly transparent conductive films based on CNT dispersions have been demonstrated, achieving a performance comparable to most of today’s commercial sources of sputtered ITO.

Moreover, the CNT films exhibit high mechanical reliability compared with ITO and can be formed using low-temperature printing techniques, making them an attractive alternative for applications requiring much lower fabrication costs and much greater flexibility [12].

In this paper, we combine, for the first time, these alternative approaches and report the incorporation of plasmonic Au NPs of polymer–fullerene photovoltaic devices, which utilize transparent single-walled CNT (SWNT) films as the hole-collecting electrode. The NP incorporation leads to an improvement of the energy conversion efficiency; this effect is attributed to an enhancement of the solar-light harvesting of the device due to the excitation of an LSPR resonance through the resonant interaction between the electromagnetic field of the incident light and the surface electron density surrounding the NPs.

II. EXPERIMENTAL

The generation of the Au NPs was performed by the ultrafast laser ablation of Au metallic target (99.99%). This technique provides the possibility of generating a large variety of NPs

that are free of both surface-active substances and counterions [14]. The targets were placed into a Pyrex cell and covered by a layer of absolute ethanol. A femtosecond (~ 100 fs at 1 kHz) laser beam was focused onto the target through the ethanol layer. The cell was mounted on a computer-driven XY stage and translated during laser exposure. More experimental details can be found elsewhere [15]. Laser irradiation gives rise to a high-temperature gradient in the metal bulk and melts of a thin layer of the target. A fraction of the molten layer of the target is dispersed into the liquid as NPs. The morphology of NPs was characterized by high-resolution field-emission transmission electron microscopy (HRTEM) performed on a Jeol JEM 2100F ultrahigh resolution at 200 kV equipped with a field-emission electron source. The size distribution of Au NPs was determined by the corresponding TEM pictures. The optical absorption spectra of the different colloidal solutions were recorded in the range of 250–1000 nm with the help of a Perkin–Elmer spectrophotometer.

The SWNT thin films were deposited on glass substrates following the method of Wu *et al.* [16]. Powders of CNTs (Carbon Nanotechnologies, Inc.) were dissolved in a 0.5% solution of sodium dodecylsulfate surfactant since the addition of the surfactant improves the solubility of the SWNTs by sidewall functionalization. The solution was bath-sonicated for 12 h and centrifuged for 60 min in order to remove metal catalysts, undispersed particles, and large nanotube bundles. Then, a controlled volume of the nanotube suspension was filtered using a 200-nm mixed cellulose ester (MCE) membrane. The filter was then attached to a glass substrate, and subsequent acetone baths were used to dissolve the MCE membrane, leaving behind the vacuum-filtered SWNT thin films [8]. The SWNT-film thickness was directly dependent on the initial volume of the SWNT suspension filtered. In order to remove the surfactant from the SWNT surface, distilled water was vacuum-filtered through the SWNT film. The thickness of the SWNT film used in this investigation was 40 nm, with a sheet resistance of $820 \Omega/\text{sq}$ and transparency of 78% at 520 nm, as previously reported by our group [10].

Regioregular P3HT and PCBM in 1 : 1 ratio were dissolved in chloroform and stirred for 1 h at 60°C . The photovoltaic devices reported were fabricated on $20 \text{ mm} \times 20 \text{ mm}$ SWNT/glass substrates. As a buffer layer, poly(ethylene-dioxythiophene) doped with poly(4-styrenesulfonate) (PEDOT:PSS) was spin-cast on the SWNT mat, followed by 3-h baking at 100°C inside a nitrogen-filled glove box. Since the LSPR involves an evanescent wave that exponentially decays with distance from the metal surface, the thickness of PEDOT:PSS was controlled to be ~ 15 nm (surface resistance $< 80 \Omega/\text{sq}$), considering the size of Au NPs. The atomic-force-microscopy (AFM) imaging of the SWNT–PEDOT:PSS film layer exhibits a low roughness of 3.0 nm after the PEDOT:PSS spin-casting, which is comparable to that of standard ITO films, which is 2.4 nm [17]. Then, the Au-NP solution (30% volume ratio) in ethanol was spin-coated on the freshly prepared PEDOT:PSS layer (see Fig. 1). The presence of Au NPs on the top of the PEDOT:PSS layer was confirmed by the AFM imaging and the scanning-electron-microscope energy-dispersive X-ray mapping (not shown) of the layer after the deposition of the NPs.

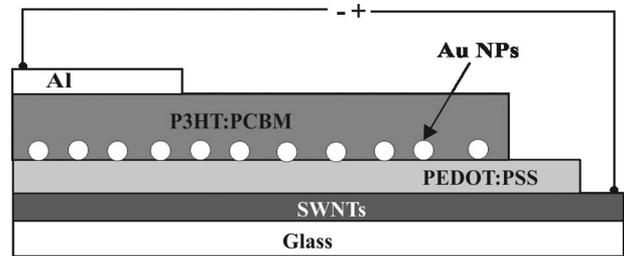


Fig. 1. Schematic of the photovoltaic device under investigation.

The photoactive layer was subsequently deposited by spin-coating the P3HT:PCBM mixture at 1000 r/min for 60 s, resulting in films with thicknesses of 200 nm. The films were dried for 1 h at room temperature and then annealed for another 30 min at 80°C in nitrogen. Aluminum cathodes were finally thermally evaporated through a shadow mask. A postfabrication annealing was performed at 110°C for 10 min in nitrogen.

The NPs were chosen to be placed below the photoactive layer rather than above the SWNT films in order to enhance the exciton yield at the interface between the photoactive layer and the PEDOT:PSS layer, due to the excitation of localized plasmon resonances in the vicinity of NPs. In this way, the excited holes are readily collected, after exciton dissociation, by the ITO electrode.

Current–voltage (I – V) measurements were performed at room temperature using an Agilent B1500A Semiconductor Device Analyzer. For the photovoltaic characterization, the devices were illuminated with a $100\text{-mW}/\text{cm}^2$ power intensity of white light by an Oriel solar simulator (active area $\sim 5 \text{ mm}^2$) with an AM1.5 filter through the glass/ITO side. Incident photon-to-current conversion efficiency (IPCE) spectra are recorded by monitoring the short-circuit current of the photovoltaic devices with a lock-in amplifier using the chopped monochromated light from a Xe lamp as illumination. All measurements were made in air immediately after the device fabrication.

III. RESULTS AND DISCUSSION

The TEM image and the corresponding size distributions of the Au NPs produced by femtosecond laser ablation are shown in Figs. 2 and 3, respectively. A dispersity in the NP sizes is observed ranging from 1.5 to 20 nm with the most-probable size being at ~ 10 nm.

Fig. 4 displays the absorption spectrum of the Au NPs in ethanol solution. The colloidal solution exhibit a distinct peak at 530 nm, corresponding to the theoretically predicted enhanced absorption due to the plasmon resonance. All the colloids prepared were stable against sedimentation for at least several months with no addition of any surface-active substances.

Fig. 5 presents the illuminated I – V characteristics of a SWNT/PEDOT:PSS/NP/P3HT:PCBM/Al configuration (as shown in Fig. 1) in comparison with a reference photovoltaic cell without Au NPs. The rectification ratio of both devices in the dark at 2 V is around 10^4 , indicating a distinct diode behavior. The photovoltaic characteristics of both devices, extracted from Fig. 5, are given in Table I.

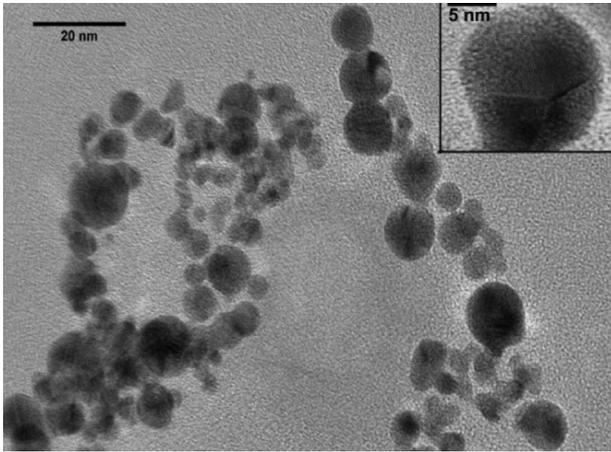


Fig. 2. TEM image of the NPs generated via the ablation of a bulk Au target in ethanol using femtosecond laser radiation. (Inset) HRTEM picture of an NP.

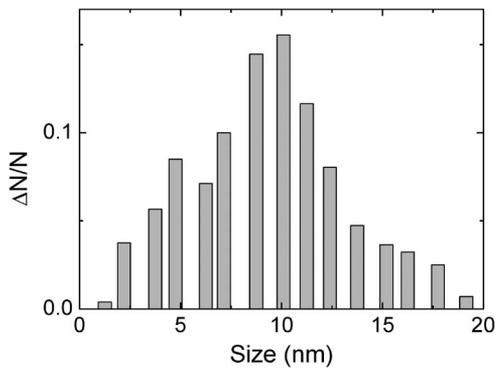


Fig. 3. Distribution of Au-NP size calculated from TEM images.

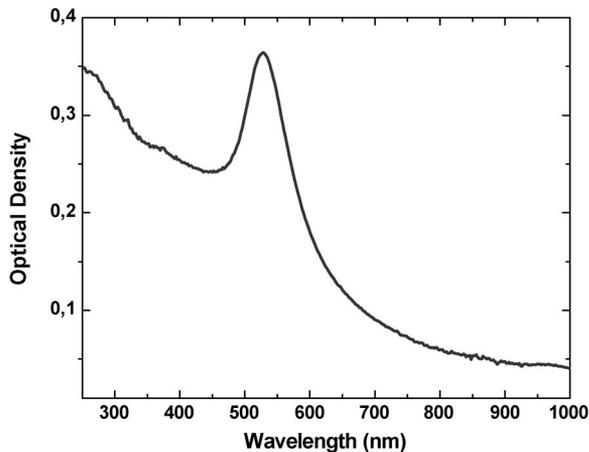


Fig. 4. Absorption spectrum of the colloidal solution of Au NPs used.

As can be seen, the introduction of the Au NPs as an interfacial layer induces a significant improvement of the device short-circuit current (by more than 30%), the fill factor (FF; by more than 20%), and the open-circuit voltage (5%). As a result, a 70% increase in the device efficiency can be obtained with the introduction of the metal NPs. It is important here to note that all devices were fabricated in air without the use of inert atmosphere; this is the reason for the low efficiency values, which, of course, are expected to be improved when working at a controlled atmosphere.

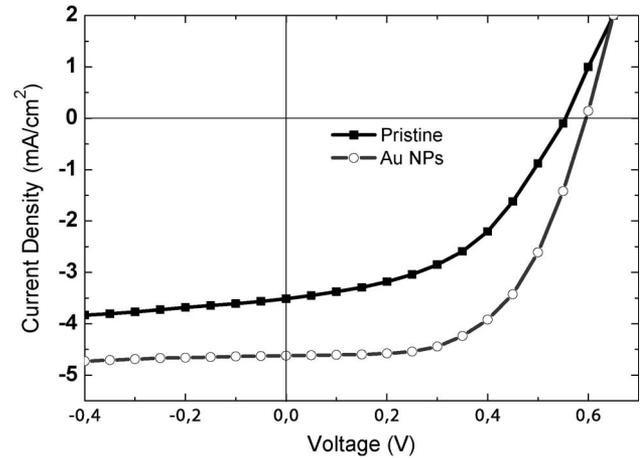


Fig. 5. Current-density-voltage (J - V) curves of the investigated photovoltaic devices under 100-mW/cm² 1.5-solar-simulator illumination.

TABLE I
PHOTOVOLTAIC PROPERTIES

| Device Structure | J_{sc} (mA/cm ²) | V_{oc} (V) | FF | η (%) |
|----------------------------------|--------------------------------|--------------|------|------------|
| SWNTs/PEDOT:PSS/NPs/P3HT:PCBM/Al | 4,6 | 0,58 | 0,58 | 1,54 |
| SWNTs/PEDOT:PSS/P3HT:PCBM/Al | 3,5 | 0,55 | 0,47 | 0,9 |

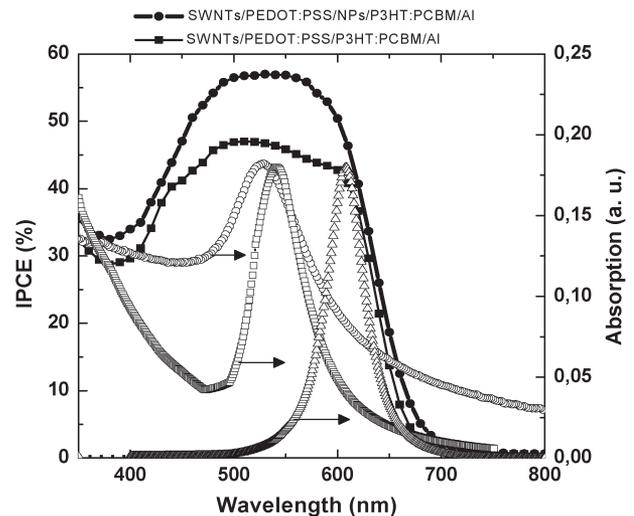


Fig. 6. (Solid symbols) IPCE of the investigated devices as a function of the wavelength of monochromatic irradiation. (Open circles) The absorption spectra of the Au NPs in ethanol and those predicted by the Mie theory for Au NPs (open squares) in PEDOT:PSS and (open triangles) in P3HT:PCBM are also presented.

In order to investigate the underlying mechanism responsible to the enhanced performance of the devices, we have measured the IPCE curves of the device with the Au NPs and have compared them with the reference cell. The IPCE is defined as the percentage of electrons, measured under short-circuit current conditions that are related to the number of incident photons. It is used to obtain information on the numbers of photons of different energies that contribute to charge generation in the device. As shown Fig. 6, within the wavelength range of 450–600 nm, the photocurrent remarkably increased upon the incorporation the Au NPs.

The observed improvement can be attributed to the enhancement in the number and/or the rate of the photogenerated excitons [17] near the metal surface. This may be indicated by the fact that an enhanced optical absorption in the range of 350–650 nm is observed for the P3HT:PCBM layers with NPs compared with that without them [19]. Metal NPs are strong scatterers of light at wavelengths near the plasmon resonance, which is due to a collective oscillation of the conduction electrons in the metal. For particles with diameters well below the wavelength of light, the scattering and absorption cross sections are given by [20]

$$\sigma_{sc} = \frac{1}{6\pi} \left(\frac{2\pi}{\lambda} \right)^4 |a|^2 \quad \sigma_{abs} = \frac{2\pi}{\lambda} Im[a]$$

where $a = 3V((\epsilon_p/\epsilon_m - 1)/(\epsilon_p/\epsilon_m + 1))$ is the polarizability of the particle. Here, V is the particle volume, ϵ_p is the dielectric function of the particle, and ϵ_m is the dielectric function of the embedding medium. We can see that, when $\epsilon_p = -2\epsilon_m$, the particle polarizability will become very large, and this is known as the LSPR. At the LSPR, the scattering cross section can well exceed the geometrical cross section of the particle.

Fig. 6 shows the extinction (scattering plus absorption) cross sections for Au spheres embedded in PEDOT:PSS and P3HT:PCBM media and calculated using the Mie theory [21], [22] and the experimental data of Fig. 3 as input for the particle size distribution. The respective dielectric constants for the two different media were obtained from [23]. For each embedding medium, a dipole resonance is observed, peaking at 540 nm for PEDOT:PSS and 610 nm for P3HT:PCBM. It is important that the maximum of the IPCE spectral-response enhancement extends to a broader wavelength range than that defined by the excitation peak of Au NPs. This may be due to the fact that the NP layer is positioned between the PEDOT:PSS and the active P3HT:PCBM layer and the observed improvement is the collective effect of the two respective resonances, which is predicted by the Mie Theory, as shown in Fig. 6.

The IPCE spectra does not provide a clear indication that the photovoltaic performance is due to an absorption enhancement effect, and therefore, the improved photocurrent may be attributed to the excitation of the LSPR. On top of that, the Au NPs may be acting as extended 3-D hole collectors in the active layer and may not be really participating in any resonance phenomena that lead to additional carrier generation. In order to exclude this option, a photovoltaic device with the same configuration but using Al NPs with a similar size distribution instead of Au ones was fabricated in identical conditions. The performance of the device incorporating Al NPs was measured to be not superior than that of the devices without NPs, indicating that the major enhancement effect is related to the enhanced absorption properties of Au NPs. Nevertheless, further work is ongoing in order to clarify this effect. The improvement in the FF can be due to the increased exciton dissociation probability resulting from the local enhancement in the electromagnetic field originating from the excitation of the LSPR [5], whereas the slight increase in the open-circuit voltage can be attributed to the change in the band alignment caused by the introduction of the Au NPs, as a similar effect is seen with ZnO NPs [24].

IV. CONCLUSION

In conclusion, the efficiency of the organic photovoltaic devices upon the introduction of Au NPs has been significantly improved. The observed improvement has been attributed to the unique optical properties of the LSPR induced by the NPs, which led to a clear enhancement of the photocurrent and the FF. This approach used in this paper is quite simple and may be suitable for the integration in roll-to-roll fabrication for organic photovoltaics. Nevertheless, many parameters still need to be optimized, including the Au NPs' density and size distribution.

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Emmanuel Kymakis (M'05) was born in Heraklion, Greece, in 1977. He received the B.Eng. (first class) degree in electrical engineering and electronics from Liverpool University, Liverpool, U.K., in 1999 and the Ph.D. degree in engineering from Cambridge University, Cambridge, U.K., in 2003. His Ph.D. dissertation (under the direction of Prof. G. A. J. Amarunga) was entitled "Organic Photovoltaic Devices Based on Conjugated Polymers and Carbon Nanotubes."

He is an Assistant Professor of electrical engineering with the Technological Educational Institute of Crete, Crete, Greece. He is also a Technical Consultant in the design and the implementation of grid-connected photovoltaic and solar thermal power plants. He was the first worldwide to suggest that carbon nanotubes can be used as electron acceptors in organic photovoltaic devices. He has over 30 publications with 1200 citations. His research interests are predominantly in the areas of novel materials and nanostructured composites, and their incorporation in optoelectronic devices, such as photovoltaic devices and field-emission displays. Furthermore, his current recent research efforts are concentrated in the investigation of nanotubes and graphene, and their unique interactions with polymers for energy conversion systems.

Dr. Kymakis is member of the Institution of Engineering and Technology and the Technical Chamber of Greece.



Emmanuel Stratakis was born in Heraklion, Greece, in 1973. He received the Ph.D. degree from the University of Crete, Heraklion, in 2001.

Since 2003, he has been a Visiting Assistant Professor with the Department of Materials Science and Technology, University of Crete, and a Research Associate with the Institute of Electronic Structure and Laser, Foundation of Research and Technology–Hellas (IESL-FORTH), Heraklion. In the autumns of 2006 and 2008, he was invited as a Visiting Researcher with the University of California, Berkeley. Since 2007, he has been a Researcher with IESL-FORTH, where he is leading the "ultrafast laser 3-D micro/nano processing" activity. He has a long-term experience in the structural, electrical, and optical characterization of materials and the development of innovative manufacturing techniques of micro/nano structures based on lasers. His scientific work includes 35 publications in refereed journals and three chapters in refereed books, and he is the holder of one patent. He currently participates in four European-Union- and Greek-administered research projects. His main research interests include the development of novel electronic structures for field emission and photovoltaic applications, the fabrication and the study of biomimetic and responsive materials, and the development of innovative diagnostic techniques at nanoscale based on atomic-force microscopy.

Dr. Stratakis is member of the European Physical Society.



Emmanuel Koudoumas was born in Heraklion, Greece, in 1960. He received the B.S. degree in physics from the National and Kapodistrian University of Athens, Athens, Greece, in 1983, the M.Sc. degree in laser and application from the University of Essex, Colchester, U.K., in 1984, the M.Sc. degree in applied physics from the University of Crete, Heraklion, in 1987, and the Ph.D. degree in physics from the University of Patras, Patras, Greece, in 1991.

From 1993 to 2002, he was a Researcher with the Institute of Electronic Structure and Lasers, Foundation of Research and Technology–Hellas. Since 2002, he has been a Professor of electrical engineering with the Technological Educational Institute of Crete, Crete. His research background includes high-intensity short-pulse interactions in atoms and molecules, nonlinear optical response of novel materials, laser-based diagnostic techniques, and generation of vacuum-ultraviolet radiation. He has over 70 publications with 700 citations. His recent research efforts are concentrated in the field of growth/characterization of novel material, such as metal-oxide nanostructures and carbon allotropes, and their use in photonic applications.



Costas Fotakis received the B.S. degree in Chemistry from the National and Kapodistrian University of Athens, in 1973 and the Ph.D. degree from the Department of the Physical Chemistry, University of Edinburgh, in 1978.

From 2005 to 2006, he was a Springer Professor with the University of California, Berkeley. He is the Director of the Institute of Electronic Structure and Laser, Foundation of Research and Technology–Hellas (IESL-FORTH), Heraklion, Greece, and a Professor of physics with the University of Crete, Heraklion. He is also the Director of the European Ultraviolet Laser Facility operating at FORTH, which is currently a part of the European Union (EU) project "LASERLAB," linking 17 major European laser infrastructures. He has been the chair or the cochair of several major international scientific conferences and member of several EU panels, including the European Strategy Forum for Research Infrastructures. He has over 200 publications primarily in the field of photonics and their applications and belongs to the editorial boards of several international scientific journals. His research interests include laser spectroscopy, molecular photophysics, laser interactions with materials, and related applications for material processing and analysis.

Prof. Fotakis is a Life Member and a Fellow of the OSA, and a member of the Fellows Committee of the European Optical Society. He is the 2004 recipient of the "Leadership Award/New Focus Prize" of the Optical Society of America (OSA) "for decade-long leadership of and personal research contribution to the field of laser applications to art conservation and leadership in establishing and guiding the scientific excellence of the laser science programs at IESL-FORTH."