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Introduction to Laser-Induced Transfer and Other Associated Processes

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Digital manufacturing emerges as a new production paradigm that is transforming the future of the manufacturing industry on both the small and the large scale [1]. Despite that this new fabrication methodology is still far from being completely consolidated, there is little doubt now that the trend toward digitization is unstoppable and that almost no industrial field will remain immune to its influence in the next years. Thanks to the capabilities of the present-day computers and CAD software, as well as to unprecedented advances in serial production technologies, digital manufacturing makes possible a truly fast transition from an idea to its realization. This integrated approach provides in turn a new concept with a high degree of flexibility in that it allows to easily incorporate design changes into the final product. All these benefits have enabled digital manufacturing in such diverse industries as electronics, energy harvesting, packaging, decoration, textile manufacturing, medical instrumentation, or regenerative medicine.

Most digital manufacturing technologies are in fact "direct-writing" technologies, that is, they allow processing materials in a serial manner following a pattern previously recorded as a digital file [2]. The direct-writing approach makes patterning possible with high resolution and control in the flexible way just described. These technologies can be either subtractive, such as laser and water jet cutting, or additive, such as stereolithography and inkjet printing. Furthermore, they are sometimes assisted by other techniques that, instead of removing or adding new material, simply modify some of the properties of an already manufactured product, also in a digital way; laser sintering and laser curing are good examples of this.

1.1 LIFT and Its Derivatives

"Laser printing" is a quite general term that applies to a rather large set of additive direct-writing techniques whose principle of operation relies on a laser-induced

Laser Printing of Functional Materials: 3D Microfabrication, Electronics and Biomedicine, First Edition. Edited by Alberto Piqué and Pere Serra.

transfer process: a controlled amount of material is transferred from a donor system to a receiving substrate by means of laser irradiation [3]. The relative displacement of the laser beam with respect to the receiver (and eventually the donor) makes patterning possible. The most common laser printing approach is known as laser-induced forward transfer (LIFT), a technique that works according to the described principle of operation with the donor system being a thin film and the laser source usually a pulsed laser. In more detail, an optical system focuses a pulsed laser beam onto a thin film of the donor material through its supporting substrate, which needs to be transparent to the laser radiation. Under the influence of a laser pulse, a tiny portion of the donor material is ejected toward the receiving substrate as sketched in Figure 1.1, which results in the formation of a pixel/voxel on that substrate. Through the repetition of this process at different positions in the donor/receiver system, any pattern can be produced.

The donor material in LIFT can be either solid [4, 5] or liquid [6, 7]. In the earliest versions of the technique (some of which date as far back as the mid-1960s [8–10]), the donor material was always solid. In those instances, transfers took place in the gas phase with the irradiated material completely vaporized [11] or in the liquid phase if melted (totally or partially). Accordingly, deposition occurred in the receiving substrate through recondensation of the vapor in the first case and through resolidification of the molten voxel in the second [4, 5]. Those transfer modes were especially suited for the printing of metals, for which extremely high resolutions were achieved [12]. However, it was found out later that transfer from solid films was also possible in the solid state, that is, without any phase change of the donor material. In that case, transfer proceeded through the laser-induced ejection of a “flyer” that was projected away from the donor film and toward the receiving substrate, where it landed, giving way to the formation of a voxel. That transfer mechanism not only allowed printing

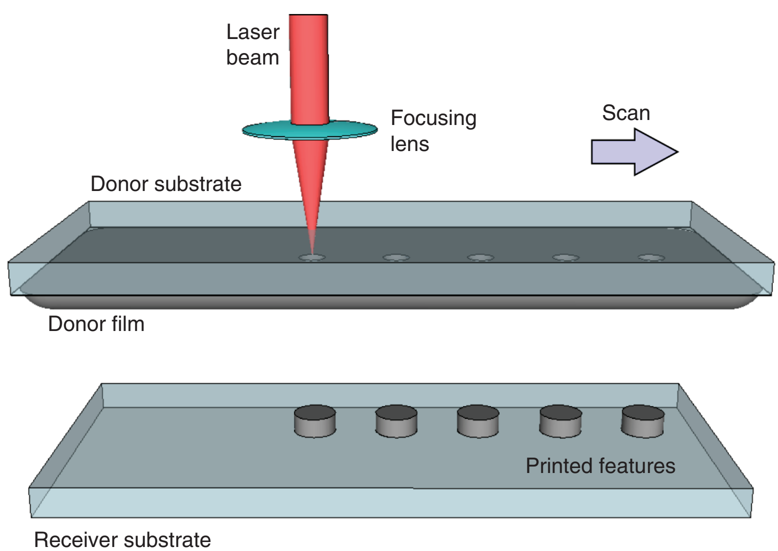

Figure 1.1 Principle of operation of the LIFT technique.

materials that would irreversibly change or decompose if melted or vaporized, such as complex ceramics [13], but also made possible to transfer stacks of different materials [14], especially interesting for the fabrication of multilayered devices. Even more than that, entire components such as surface-mounted devices and bare dies can be successfully transferred through LIFT [15].

LIFT can also work with liquid donor films. In this case, the material to print is previously dissolved or suspended in an ink that is transferred as a whole. The principle of operation of the technique is the same as with solid donor films (Figure 1.1), with the printing outcome now being a sessile droplet of the ink. Further evaporation of the ink solvents leads to the formation of the resulting pixel on the receiving substrate. The main difference with the LIFT of solids relies on the transfer mechanism: the ink is projected away from the donor film through a high-speed liquid jet that results from the expansion and collapse of a laser-induced cavitation bubble [16]. The LIFT of liquids is remarkably similar to inkjet printing regarding both transfer mechanism and printing outcome, but with significant advantages over it. Probably the most prominent is that LIFT presents fewer restrictions concerning the rheology of the printable inks: LIFT admits a substantially broader range of viscosities [17] and sizes of the particles loading the ink [18] compared to inkjet printing. Thus, such technologically interesting materials as high-solid-content inks and nanostructured materials can be successfully printed through LIFT. Furthermore, since those transfers also proceed without any significant phase change of the printed material, LIFT from liquid donor films can be used to print very sensitive materials such as biomolecules [19, 20] or living cells and tissue [21–23], which makes the technique especially suited for biosensing and tissue engineering applications.

In spite of its dominant position in the laser printing techniques universe, LIFT is not the only option to carry out laser-induced transfer. One alternative is laser-induced backward transfer (LIBT), wherein the donor film is irradiated directly, either at normal incidence through a transparent receiving substrate [2, 24] or at oblique incidence with flexible receiving substrates [25], and the irradiated material is propelled in a direction opposite to that of the incident laser beam. This approach is more restrictive and probably more difficult to set up compared to LIFT, but it has proved to be very convenient for the large-area and large-throughput printing of liquid inks [25]. Another novel departure from the traditional LIFT scheme is usually known as film-free laser printing [26]. This approach, which works only with liquid inks transparent to the laser radiation, requires the use of very short laser pulses. However, it presents the advantage of allowing to carry out transfers directly from the ink contained in a reservoir, without the need of preparing a thin film. Film-free laser printing can operate in both forward and backward configurations.

1.2 The Laser Transfer Universe

The concept of imparting kinetic energy to a solid using laser pulses can be traced back to research on the use of lasers for rocket propulsion applications and launching of thin films to study matter under the effects of hypervelocities (>1 km/s) [27]. Early studies in the 1970s revealed that the plasma generated during the laser-induced breakdown of air near a surface would impinge on and transfer momentum to the surface [28, 29]. The use of pulsed lasers to accelerate small objects such as thin films has since then been explored to investigate means for propulsion of micro-rockets [30] and the effects of micrometeorites to simulate the effects of space debris on satellites [31–33]. These works focused on the efficiency of the release and launch process but share many similarities with LIFT.

The straight forwardness of the laser release and launch process and its relative simplicity in implementation attracted much attention in the graphics industry for digital printing tools [10]. In fact, Levene *et al*. reported in 1970 the laser-induced transfer of material across an air gap for the purposes of printing characters, recording of graphics, or marking applications [34]. The material transferred consisted of black ink from a polyethylene-backed typewriter ribbon and colored dies from a Mylar substrate across up to 100 μm gaps using a Nd:YAG laser ($\lambda = 1.06$ μm). Although the objective of this work was to develop a laser-based printing or marking process (the authors referred to it as material transfer recording), it was prescient in many ways including its reporting of not only forward but also backward transfers (called reverse transfer by the authors) and on proposing a simple physical model based on the melting and vaporization of the transferred material as a function of the laser pulse energy. Unfortunately, the authors did not apply their technique to any other types of materials, and their work went unnoticed until the late 1990s when their article began being cited within the printing and image science community. The laser forward-transfer process had to wait until 1986 to be rediscovered, this time with metals, with the work by Bohandy *et al*., who reported on the deposition of copper metal patterns using pulses from a UV laser ($\lambda = 193$ nm) and proposed a physical model more detailed but similar to Levene's to describe the process [4]. Furthermore, it was Bohandy's group who coined the term LIFT (for laser-induced forward transfer) to denote the process, which is essentially the subject of this book.

It was with LIFT that the use of lasers to achieve the controlled transfer of a specific material from a donor to a receiving substrate became widespread to the point that the acronym itself began to be used by some as a verb describing the process as in "LIFT-ing" a given material or ink. The success of LIFT also gave rise to numerous derivatives, all laser-based and all with their own acronym aiming at differentiating themselves from the basic technique. This book does not include all these LIFT derivatives since their discussion is beyond the scope of this volume. However, the remaining of this Introduction will provide a brief examination of some of these laser transfer techniques and include those relevant references where the reader will be able to find more details.

Early on, applications in the graphics industry motivated the use of laser transfer processes for image recording as evidenced by Levene's work. In these applications, the transfer process involves the conversion of light into heat when IR laser pulses are absorbed by IR-absorbing dye pigments dispersed in a translucent binder. Ultrafast heating of the absorbing dyes causes their decomposition, vapor generation, and thermal stresses leading to thermal ablation. These processes known as laser ablative transfer or LAT involve delamination, rupture, and launch of the film from its carrier when the pressure of the generated gases trapped in the absorbing layer exceeds a threshold determined by the nature and thickness of the absorbing layer and laser parameters, that is, wavelength, pulse width, fluence, and so on. With LAT, it is possible to print high-resolution full-color graphics using a dry, noncontact printing process. Analysis of the LAT process was undertaken in the 1990s to determine its mechanism and optimize its operation by D. Dlott's group at the University of Illinois at Urbana Champaign. They reported various key findings including measuring heating rates and peak temperatures using dyes as molecular thermometers [35, 36] and applying ultrafast microscopy of shock waves generated with ps laser pulses [37, 38]. LAT can also be used to print nonabsorbing layers by incorporating a sacrificial laser absorbing layer first referred by Dlott's group as the dynamic release layer or DRL [39]. Finally, using a metal DRL and ps laser pulses, LAT can be achieved at low fluences ($<10\,mJ/cm^2$) by vaporizing the thin aluminum interlayer instead of the dyes, generating color images at ultrahigh speed and high resolution [40].

Another class of laser transfer techniques owe their development to the need to find alternatives for the generation of patterns, given the cost and complexity of conventional vacuum deposition and photolithography-based process. In many instances, these laser transfer derivatives were developed to address a specific class of material(s) for a given application as in the case of the organic semiconductor molecules used for organic electronic devices and organic light-emitting diodes (OLEDs). These materials present serious challenges when applied as liquid layers for large-area printing and would benefit from a dry printing process. These needs have given rise to several techniques based on laser transfer, which have meet with various levels of success. One of these techniques known as laser-induced thermal imaging or LITI was originally developed at DuPont by Blanchet's group for printing conducting polymers for organic electronics [41]. In LITI, the precoated donor and receiving films are in contact and held together using vacuum. A IR cw laser is aimed through the donor film at a thin absorbing layer for conversion of the light into heat, which vaporizes an intermediate organic layer with the resulting gas implanting a third layer comprising the conductive polymer onto the receiving film. This thermal transfer method relies on release/adhesion mechanisms rather than ablation of the conductive polymer (or organic semiconductor) layers, allowing them to remain continuous rather than undergoing irreversible damage (as with ablation). LITI was further developed by the 3 M Corporation for the fabrication of LCD color filters and OLED emitters [42], and beginning in 2000, 3 M partnered with Samsung SDI to jointly develop the process for active-matrix organic light-emitting diode (AMOLED) displays [43]. The use of LITI in the fabrication of AMOLED displays, which are commonly used in smartphones, represents the most successful example to date of the commercial application of laser transfer techniques. Interesting enough, given the commercial success of LITI, other consumer product companies have developed their own versions of laser transfer technologies such as laser-induced patternwise sublimation or LIPS, which has been developed by Sony Corporation. Using LIPS, RGB pixels are patterned for OLED applications, which have been used by the Display Device

Development group at Sony Corporation to fabricate a 27.3-inch AMOLED display [44]. For heat-sensitive organic materials, LITI and LIPS might cause their damage, and the use of more efficient light-absorbing layers based on loosely connected silver nanoparticles has been investigated. This technique named nanomaterial-enabled laser transfer or NELT allows the laser printing of patterns of the OLED tris-(8-hydroxyquinoline)Al, better known as Alq_3, without damage [45]. These laser transfer techniques for organic electronics are highly proprietary, each being the subject of numerous patents, and require the use of multilayer, complex, and, in most cases, proprietary donor films, which has limited their use by the general LIFT community.

The implantation of intact polymer molecules using a laser transfer process has been reported by H. Fukumura's group from Tohuku University. The technique known as laser molecular implantation or LMI is used for the implantation of fluorescent molecules into polymer films at precise locations and depths [46]. In LMI, the laser passes through a transparent polymer serving as the receiving substrate, such as poly(butyl methacrylate) or PBMA, or poly(ethyl methacrylate) or PEMA, which is in contact with an absorbing polymer film containing the organic molecules to be implanted, such as pyrene [47] or photochromic molecules [48]. More recently, there have been reports on laser-implanted coumarin C545 with a process called laser-induced molecular implantation technique (LIMIT) for organic electronics and optical device applications [49]. Both LMI and LIMIT are very similar, and their use is limited to applications requiring the implantation of specific molecules on a polymer matrix just below its surface, and as such, they have not found widespread use.

As a last example of laser-mediated transfers of a material or sample, it is worth considering the process used for laser pressure catapulting or LPC. This process has been used to procure single cells or small regions comprised of homogeneous cells for subsequent molecular analysis using UV laser pulses [50]. LPC is a noncontact cell or tissue capture technique that in combination with microdissection allows the rapid preparation of samples for cell or tissue analysis. LPC can also be used for the collection of chromosomes and chromosome subfragments for gene research applications [51]. Later reports have demonstrated the use of LPC for the transfer of living cells from a microdissected cell culture, which are viable enough to be recultivated [52, 53]. The drawback of LPC is that it requires a donor film prepared by microdissection, which, given their limited size, prevents this technique from its use in transferring much larger numbers of cells as for tissue engineering applications.

1.3 Book Organization and Chapter Overview

The aim of this book is to provide an extensive overview of the topic of laser printing of functional materials. As pointed out in the preceding sections, the editors are aware that the 18 chapters of this book have not covered all the existing laser-induced transfer techniques. Instead of attempting to cover the entire field, we have preferred to focus on LIFT and its most closely related technologies, paying special attention to their principles of operation and transfer mechanisms, without forgetting their potential and applications. In other words, we have carefully selected the most popular approaches to laser printing and aimed to provide an in-depth treatment of all of them. This book is the result of contributions by international leading experts in the field, both from the academia and from the industry, thus providing a broad perspective of the present and future of these technologies.

This book is organized into three main parts. The first one deals with the fundamentals of laser printing. In this part, the main techniques are presented and described in detail, alongside the mechanisms of transfer involved in each of them, but with no special focus on any material in particular.

In Chapter 2, by C. Kryou and I. Zergioti, the authors provide an overview of the main laser-induced transfer processes and their historical origin. This chapter serves as an introduction to the techniques that will be later analyzed in more detail, outlining their main characteristics as well as some important instrumental issues. In this chapter, the authors also introduce some of the approaches to laser printing away from the mainstream. Since these approaches are not considered in the rest of this book, by presenting them here, the authors provide the interested reader with the appropriate references for further investigation.

We devote Chapter 3, by A. Palla Papavlu and T. Lippert, to a common variation to traditional LIFT: the use of a DRL. The transfer of very sensitive materials, as well as materials transparent to the laser radiation, is usually carried out through an intermediate absorbing film. The laser pulse is absorbed within that film, which provides the donor material with the thrust required for transfer without the sensitive material being exposed to the potentially harmful laser radiation. In this chapter, the authors focus their discussion on the use of triazene polymers, by which absorption can be easily tuned to several convenient laser wavelengths and by which decomposition products upon laser irradiation are very efficient in propelling the donor material.

In Chapter 4, by J.M. Fernández-Pradas *et al.*, the authors introduce the general elements of the LIFT of fluids, probably the most widespread laser-based technique for printing inks. The authors describe the principle of operation of this particular approach to LIFT and discuss the mechanisms of liquid transfer, with their characteristic jetting dynamics. In this chapter, the authors also investigate the influence of the main process parameters on the morphology of the printed features and their correlation with the different possible transfer dynamics. Finally, the chapter considers some of the issues related with the printing of continuous patterns.

We return to the laser printing of fluids in Chapter 5, by E. Turkoz *et al.*, where the authors present another approach to LIFT with an intermediate layer: blister-actuated laser-induced forward transfer (BA-LIFT). In contrast to the strategy presented in Chapter 3, in this case, the laser-absorbing layer does not decompose completely. Upon absorption of the laser pulse, only a fraction of the relatively thick polymeric layer is ablated. The expansion of the trapped ablation products results in a blister in that layer, which provides the donor material with the required impulse for transfer. The mechanisms of liquid ejection are discussed and analyzed in detail both experimentally and through computational methods.

In Chapter 6, by S. Surdo *et al*., the authors depart from the LIFT scheme to present film-free laser printing (or film-free LIFT), a technique that makes possible the transfer of liquids from bulk material. After discussing the problems associated with the preparation of a thin liquid film, the authors present the principle of operation of this relatively new approach and analyze in detail the mechanisms of material transfer, with a special emphasis on the differences with traditional LIFT. In this chapter, the authors also consider important instrumental aspects, as well as the different configurations through which film-free LIFT can be carried out (forward and backward). Finally, the chapter provides some examples of applications.

The second part of this book focuses on the laser–material interaction in laser printing, emphasizing those aspects that are specific to each kind of material (metals, ceramics, etc.). This part is organized according to a general materials classification.

We start this part with a complete overview of the LIFT of metals in Chapter 7, by D.A. Willis. After a general introduction highlighting the thermal nature of metal ablation, Willis reviews the main experimental and computational studies on the mechanisms of metal transfer through LIFT and the influence of the different process parameters on them. In this chapter, special attention is paid to a particular mode of deposition apparently specific to the LIFT of metals: droplet deposition. This remarkable transfer mode has allowed printing of the smallest features ever attained through laser-based processes.

In Chapter 8, by B. Mills *et al*., the authors discuss the laser transfer of materials from solid films, an alternative particularly interesting for printing ceramics and polymers. In contrast to metals, in this case, the material is not melted, but rather transferred as a solid flyer that is propelled from the solid donor film upon absorption of the laser pulse. The authors discuss the importance of laser beam shaping in this particular approach and analyze the main issues associated with its transfer dynamics, which appears to be quite different from most of the other laser printing approaches. Although most of the chapter is devoted to LIFT, it also includes some discussion on the LIBT of nanoimprinted polymers.

Chapter 9, by Z. Zhang *et al*., discusses LIFT of soft materials. After having introduced the LIFT of fluids in Chapter 4, the focus of attention in this chapter is aimed toward non-Newtonian fluids, with a transfer dynamics sensibly different from that analyzed in the first part of this book. The authors analyze the particular jetting behavior of these fluids, with special attention to the influence of their viscoelastic properties on the dynamics. It is remarkably interesting in this case how those viscoelastic properties make possible to build 3D structures through LIFT by means of stacking successive layers of material.

In Chapter 10, by R.C.Y. Auyeung *et al*., the authors discuss LIFT of high-viscosity nanopastes, a particular type of inks that somehow combine some of the most relevant elements encountered in Chapters 8 and 9. On the one hand, a relatively soft material can be transferred, and on the other hand, as a congruent flyer. This approach, also known as laser decal transfer (LDT), indeed makes it possible to transfer complex geometries with a single laser pulse. Furthermore, it allows achieving conformal deposition, which makes it especially suited for electrical contacts, for example. Similarly to the LIFT of soft materials, 3D printing through multilayer stacks is also possible.

In Chapter 11, by U. Zywietz *et al*., the authors expand on the droplet deposition mode presented in Chapter 7 for the LIFT of metals with the aim of printing nanoparticles. In fact, the authors show that LIFT in that mode is also possible with semiconductors and demonstrate that extremely small features can indeed be obtained in that way. This chapter also explores alternatives to the traditional mode: a backward configuration for the transfer of individual nanoparticles, as well as the possibility of printing more complex patterns from prestructured films.

The last part of this book is devoted to applications. As many other digital printing techniques, laser-induced transfer can be used in a wide range of applications, some of them well consolidated in the industry and others still prospective. In this part, we present a rather comprehensive list of examples of application of laser printing and discuss the main advantages and bottlenecks of these techniques regarding their industrial implementation.

The first application that we consider is the laser printing of electronic materials, to which Chapter 12, by P. Delaporte *et al*., is devoted. Although the chapter extensively discusses the fabrication of organic devices (thin-film transistors and light-emitting diodes), the transfer of inorganic materials (dielectrics and conductive metals, especially) is also covered. The authors present examples of successful printing of electronic materials in both solid and liquid states, as well as printing of multistack structures. In addition to the feasibility of laser-induced transfer for the fabrication of electronic devices, the chapter considers issues related to the further industrialization of the technology, such as the process speed.

In Chapter 13, by I. Zergioti, the feasibility of laser printing for producing chemical and biological sensors is explored. After an overview on the most conventional printing techniques, the authors introduce LIFT of sensing materials. The chapter reviews the different approaches of laser printing for sensor fabrication, both in the solid state and in the liquid state, and with a broad variety of materials (inorganic, organic, biomolecules, nanostructured materials), to finally provide proofs of concept of functional sensors fabricated through laser-induced transfer processes.

Strongly related with the previous chapter, in Chapter 14, by A. Palla Papavlu *et al*., the authors consider the laser printing of proteins and biomaterials. Some of these materials can indeed be used for the fabrication of biosensors; however, there are many other interesting applications that require depositing biological materials with high resolution and control: cell growth and development studies, diagnostic tests, basic studies of biomolecule interactions, drug delivery, etc. The chapter discusses all these applications with a special focus on the particularities related with the laser-induced transfer of each type of biomolecule.

In Chapter 15, by O. Kérourédan *et al*., the authors make a step forward on the scale of biological complexity toward the laser printing of cells for tissue engineering applications. After an introduction to the subject of bioprinting, the authors focus their attention on the laser transfer of living cells and the influence of the diverse process parameters on cell viability and behavior. In the frame of this investigation, it is especially important to provide analysis of cell–cell and cell–microenvironment interactions, issues that the chapter discusses alongside the remarkable problem of vascularization, essential for the correct development of printed tissues. Finally, the chapter reviews some studies on the laser printing of the most relevant tissues for regenerative medicine applications.

Chapter 16, by G. Hennig *et al*., provides a complete analysis on the industrial perspectives of laser printing, with a special focus on Lasersonic®, probably the largest printing machine based on LIFT/LIBT technologies to date. In this chapter, the authors review the state of the art of laser printing technologies in the industrial arena and discuss in all detail the main instrumental issues to be considered in the setting up of an industrial-scale printing machine. This is, in consequence, the chapter with strongest engineering focus in this book, which should serve as a bridge between the research and the development stage.

Chapter 17, by R. Pohl *et al*., returns to the LIFT of metals, this time to carry out 3D printing. The ejection of extremely small metallic droplets described in Chapters 7 and 11 can be exploited to generate 3D structures. The process requires the fine-tuning of the laser parameters, as well as a detailed knowledge of the transfer dynamics, acquired through time-resolved imaging techniques. High-aspect-ratio freestanding pillars are generated through stacking successive melted droplets, which solidify immediately upon landing on the growing pillar. These elements constitute the building blocks that will be used later to produce more complex 3D structures.

The last chapter of this book, Chapter 18, by A. Piqué *et al*., is devoted to the laser transfer of entire structures and functional devices. Instead of using laser printing techniques to transfer materials with the aim of manufacturing devices, in this chapter, the authors take advantage of the principle of operation of LIFT to propel entire structures and devices. The LDT technique presented in Chapter 10 can be successfully used to print interconnects in electronic circuits, for example. But the same scheme allows the transfer of small integrated devices, such as surface-mounted devices or bare dies, which can be positioned with high accuracy and control on the required positions in a more complex circuit.

1.4 Looking Ahead

As this brief introduction shows, laser transfer techniques have evolved significantly and found widespread use in many fields. Furthermore, with the advent of LIFT, these techniques have given rise to a community of practitioners who continue investigating its mechanisms and exploring its applications. This book is an attempt to document the origin and evolution of LIFT and all its variations while providing a snapshot of the broad range of present-day applications for which LIFT provides practical solutions. We cannot predict where LIFT will take us in 5 or 10 years from now, but we hope that the discussions presented by the authors of all the chapters in this book serve to lay the groundwork for the next generation of LIFT-based techniques and their new applications.

Acknowledgments

AP would like to acknowledge the support of the Office of Naval Research (ONR) through the Naval Research Laboratory Basic Research Program.

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