

Fundamental and Applied Research in Soft Lithography at Bell Laboratories: 1997-2002

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Bell Laboratories has a long, successful history of research and innovation in lithographic methods for fabricating advanced electronic and optoelectronic devices – from early efforts on X-ray techniques, to projection-mode electron beam systems, to laser interference approaches, to proximity-corrected photolithographic schemes. Notable activities that culminated with commercial translation include electron beam exposure tools for mask making (Don Herriott and others) and proof-of-concept demonstrations in extreme ultraviolet lithography (Obert Wood, John Bjorkholm and others). These programs also involved development of photosensitive and electron beam responsive resist materials (Elsa Reichmanis, Frank Houlihan, Om Nalamasu and others).

In the late 1990's and early 2000's, this work expanded to encompass emerging strategies based on soft molds, elastomeric stamps and conformal phase masks, each with nanoscale resolution -- collectively referred to as soft lithographic techniques. Research in these areas balanced both fundamental and applied aspects, the latter pursued in a highly collaborative mode across several laboratory divisions and business units. The Condensed Matter Physics Department served as the centroid of these activities, initially through my role as a Member of Technical Staff (MTS; starting in Fall 1997) and then as Director (starting in late 2000) of the department.

The efforts adopted and extended approaches co-developed at Harvard University while I was with Professor George Whitesides as a Junior Fellow in the Harvard University Society of Fellows, just before I joined the Labs. One of the most prominent such methods is microcontact printing, as a means to pattern self-assembled monolayers (SAMs, typically alkanethiols) on surfaces (typically thin films of coinage metals) using high resolution stamps formed in soft elastomers (typically poly(dimethylsiloxane), PDMS). This class of chemistry was discovered at the Labs some years prior by Ralph Nuzzo, and was studied extensively by him and his collaborators (David Allara, Larry DuBois and others). Printed patterns of SAMs with lateral feature sizes that extend into the deep sub-micron range can be used to guide the removal or deposition of functional materials, as a complete lithographic process. Relative to traditional approaches, key unique features are in the ability to pattern (i) large areas (e.g. square meters or more) in a single step or in a roll-to-roll fashion, (ii) highly curved surfaces (e.g. optical fibers), and (iii) chemically or thermally fragile materials (e.g. organic semiconductors).

These techniques served as the basis for collaborative initiatives that we launched immediately upon my arrival at the Labs. The first involved projects with staff in the optical fiber division (Ben Eggleton, Tom Strasser, Ken Feder, Rob Windeler and others), oriented around developing components for dense wavelength division multiplexed (WDM) communication systems of core business interest to Lucent Technologies, the parent company at that time. My broad concept was to use microcontact printing to form active circuit elements on the outer surfaces of fibers, with designs that might allow fast, 'in-line' manipulation of optical signals without the need for out-coupling. Our initial work focused on simple examples in passive fiber devices, such as printed 'on-fiber' photomasks to spatially modulate the exposure to UV light, as an improved process for manufacturing in-fiber gratings as filters and reflectors [1] (with Jeff Wagener and Ashish Vengsarkar). These efforts quickly broadened to include active devices based on printed microcoils as heating elements for dynamically modulating the characteristics of in-fiber Bragg and long period gratings

and for stabilizing their properties [2-6] (with Eggleton, Todd Salamon, Alexei Abramov, Windeler, Strasser and others). This project then soon shifted to designs for producing precisely defined thermal gradients along the lengths of in-fiber Bragg gratings with intrinsic chirp, for tuning the magnitude of this chirp [7-9] (with Eggleton, Strasser, Rebecca Jackman and others). When operated in reflection mode, these systems provide dynamically adjustable levels of chromatic dispersion, with the ability to actively and adaptively cancel the detrimental effects of time-dependent dispersion observed in long-haul WDM systems with 40 Gb/s per-channel capacity, then under intense development [10-13] (with Eggleton, Benny Mikkelsen, Ashish Ahuja, Paul Westbrook, Torben Nielsen and others). Extensive prototyping, optimization and testing leveraged a broad set of collaborations spanning the physics lab and the fiber division. The outcomes led to publications, conference presentations, system level demonstrations and, ultimately, a commercial product – the RightWave Tunable Dispersion Compensator (TDC) – as an enabling component of the most advanced WDM systems offered by Lucent at that time. The technology was also later released in the form of a separate device by Furukawa Electric Co. after their acquisition of Lucent’s Optical Fiber Solutions business unit. See Figure 1.

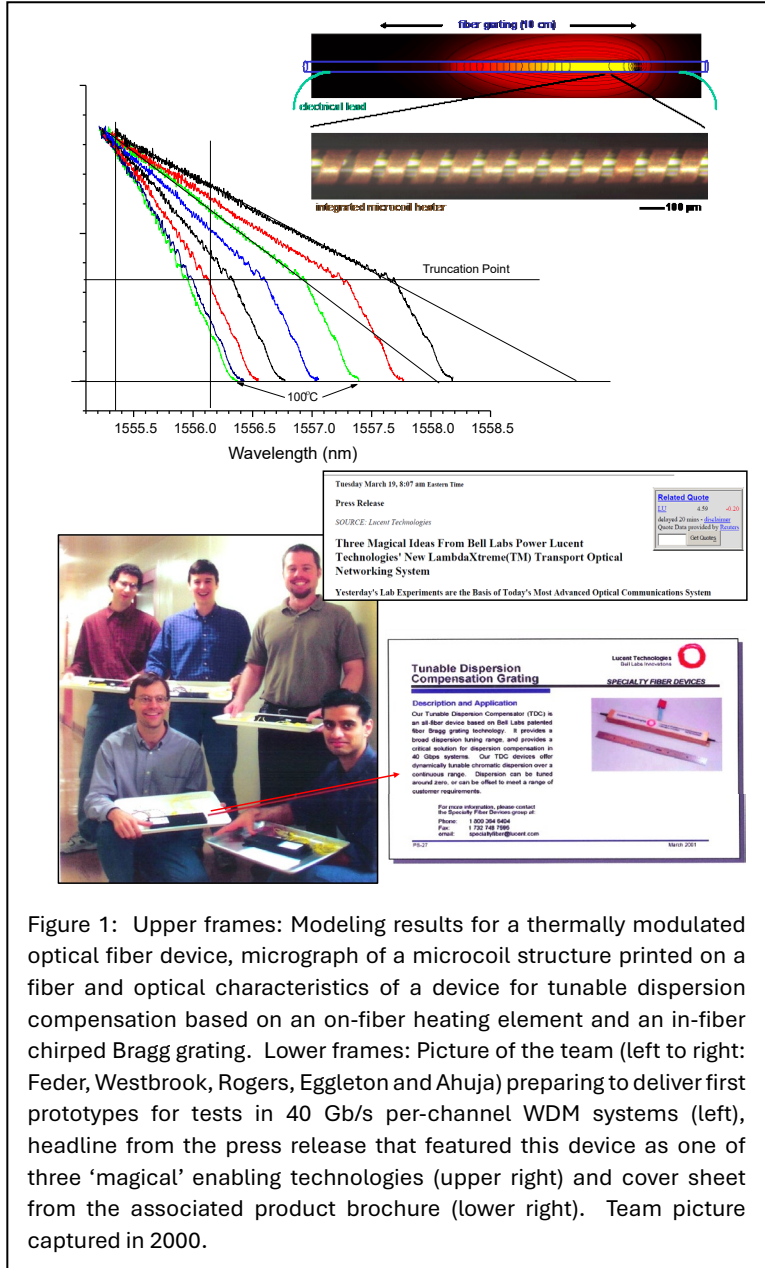


Figure 1: Upper frames: Modeling results for a thermally modulated optical fiber device, micrograph of a microcoil structure printed on a fiber and optical characteristics of a device for tunable dispersion compensation based on an on-fiber heating element and an in-fiber chirped Bragg grating. Lower frames: Picture of the team (left to right: Feder, Westbrook, Rogers, Eggleton and Ahuja) preparing to deliver first prototypes for tests in 40 Gb/s per-channel WDM systems (left), headline from the press release that featured this device as one of three ‘magical’ enabling technologies (upper right) and cover sheet from the associated product brochure (lower right). Team picture captured in 2000.

Our research in this broader area expanded to focus on integration of various ‘on-fiber’ actuators with classes of ‘holey’ fibers that were just then beginning to emerge from research on novel fiber preforms and drawing techniques. Examples included electrodes designed to modulate the properties of active materials filled into microchannel structures within these fibers [14] (with Eggleton, Ahuja, Feder, Charles Kerbage, Paul Steinvurzel, Westbrook, Windeler and others) – from polymer dispersed liquid crystals (PDLCs) electro-optically adjusted to alter losses from scattering mechanisms [15] (with Peter Mach, Kerbage, Windeler, Eggleton and others) to high index liquids

rapidly pumped back and forth along the length of the fiber to adjust essential waveguiding characteristics [16,17] (with Mach, Kerbage, Kirk Baldwin, Windeler, Eggleton and others). Realization of classes of tunable, wavelength-selective filters based on active, microfluidic fiber devices with long-period gratings in their cores represented a key achievement.

A second stream of work, pursued simultaneously with these efforts in fiber devices, exploited unique capabilities of microcontact printing in the field of organic electronics. Here, our programs involved collaboration with a collection of chemists, electrical engineers and materials scientists who had recently developed materials, circuit design approaches and screen-printing methods for building electronics on flexible plastic substrates. A key limitation at the time that I joined the team was a lack of methods to pattern these circuits with sufficiently high resolution for envisioned applications in low-cost RFID tags, flexible, ‘paper-like’ displays and others. I aimed to develop such methods and to use them in convincing product-level demonstrators. Our research focused first on simple arrays of transistors [18] and ‘smart pixels’ that integrate organic transistors with organic LEDs [19] (with Zhenan Bao, Ananth Dodabalapur and others) or PDLCs [20] (with Mach, Rob Nortrup, Pierre Wiltzius and others). We also adapted versions of microcontact printing for use with metals and ultrathin dielectric materials derived from solution-based growth techniques [21] (with Chris Jones, Don Murphy and others).

Toward the latter part of 1999, these successful outcomes led Bill Brinkman (then Vice

President of Physical Sciences Research) to challenge the group to combine our best materials with our most advanced patterning methods to build a ‘showpiece’ device capable of illustrating the promise of printed organic electronics to the broader public. The Director of my department, Pierre Wiltzius, partnered with the Director of a materials department (Reichmanis) to co-manage an effort configured to meet this challenge. Over the following ~12 months, we (with Bao, Dodabalapur, Baldwin and others) worked closely with engineers at the startup company E-Ink, Inc. to combine sheets of their electrophoretic ink (later the basis of the Amazon Kindle reader device) with thin, flexible active-matrix backplane circuits formed by our team using large-area microcontact printing. The program culminated with the production of several prototype devices – the world’s first electronic paper-like displays – including one that was demonstrated live for Bill Brinkman in his office sometime in the latter part of 2000. We published the details in 2001 in *Science* [22] and in the *Proceedings of the National Academy of Sciences* as a cover feature article [23] with a companion commentary piece

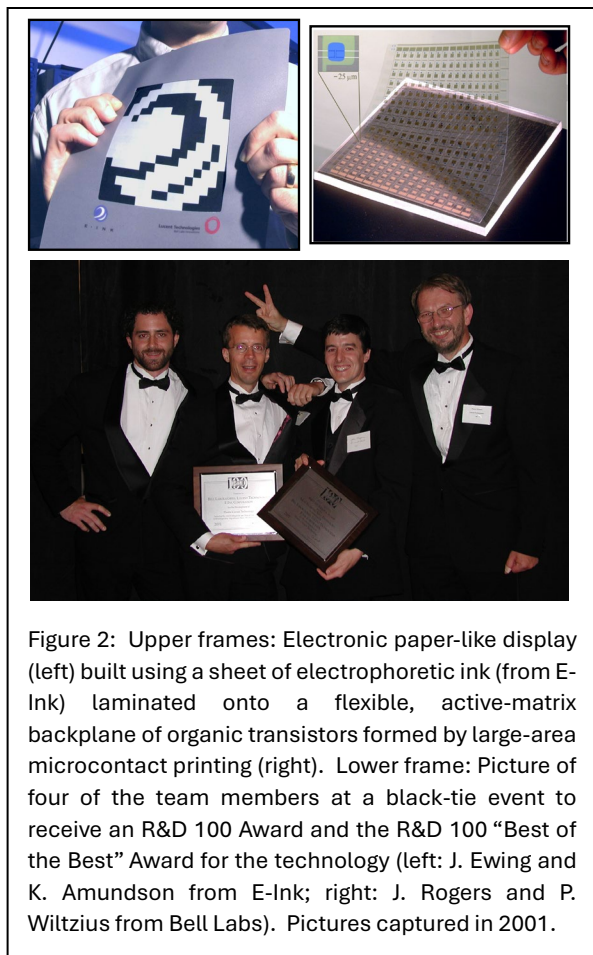


Figure 2: Upper frames: Electronic paper-like display (left) built using a sheet of electrophoretic ink (from E-Ink) laminated onto a flexible, active-matrix backplane of organic transistors formed by large-area microcontact printing (right). Lower frame: Picture of four of the team members at a black-tie event to receive an R&D 100 Award and the R&D 100 “Best of the Best” Award for the technology (left: J. Ewing and K. Amundson from E-Ink; right: J. Rogers and P. Wiltzius from Bell Labs). Pictures captured in 2001.

written by Ralph Nuzzo, then a faculty member at the University of Illinois at Urbana/Champaign [24]. These publications generated saturation coverage in the worldwide popular press. Licenses to the intellectual property created by these activities were granted to E-Ink, in exchange for an equity position in their company. See Figure 2.

Beyond these application-oriented projects, we continued our work on basic physics and materials aspects of various soft lithographic techniques and extensions of them. An important example involved the use of PDMS stamps as conformal photomasks. In a contact exposure mode, these masks serve as the basis for a type of near-field, phase-shift photolithography capable of ~100 nm resolution even with simple UV Hg lamps as light sources. We performed quantitative modeling of the optics to reveal the basic physics of the technique and we demonstrated the scheme in the fabrication of nanoscale organic transistors [25] (with Dodabalapur, Bao and others) to highlight their utility in high-speed flexible electronics.

Other initiatives focused on the stamps as soft molds for forming nanoscale structures of relief in specialized photopolymers developed for this purpose, through exploratory efforts at the wafer-scale in the cleanroom at Murray Hill, NJ and in an optoelectronic production facility at Breinigsville, PA. We fabricated a broad range of research devices using these methods, including DFB, DBR and photonic crystal laser resonators [26-29] (with Dodabalapur, Joe Timko, Dick Slusher and others), microlens arrays [30,31] (with Houlihan, Peter Mirau, Alex Liddle, Nalamasu and others) and organic LEDs [32] (with Bao, Lisa Dhar and others).

In additional work, we explored the ability to print solid material elements, rather than molecules. These activities led to techniques known as micro/nanotransfer printing, illustrated through various 2D and 3D nanostructures formed directly in thin metal films (with Lynn Loo, Dave Lang, Hsu, Bob Willett, Baldwin and others), with applications in organic transistors, photonic crystals and others [33-36]. Related examples were in 'laminated' organic LEDs, transistors and simple circuits [37-40]. Unusual test vehicles created in this way, including those based on 'transistor stamps', opened up opportunities for our fundamental studies of contact resistances [41-43] (with Loo, Lang, Hsu, Jana Zaumseil, Baldwin and others) as well as out-of-plane [39,42,43] (with Tae Woo Lee, Hsu, Zaumseil and others) and anisotropic in-plane [38] (with Zaumseil and Vikram Sundar) charge transport in organic semiconductors.

These core ideas in transfer printing soon evolved to allow manipulation of micro/nanostructures of inorganic semiconductors, including device-grade monocrystalline silicon. The mechanisms rely on reversibly modulating the physics of soft adhesion at the surfaces of the stamps by exploiting rate-dependent effects associated with viscoelastic elastomers incorporated into these systems. The advanced methods that we developed are now in broad commercial use for heterogeneous integration in optoelectronics manufacturing. In fact, they remain an area of active development at the current instantiation of Bell Labs (now owned by Nokia), highlighted most recently at a symposium held at the Murray Hill auditorium in April, 2024 to honor Louis Brus, a winner of the 2023 prize in chemistry -- the 10th Nobel Prize for work at the Labs.

My transition to Director of the department toward the end of 2000 led to an increased emphasis on corporate priorities, which included severe and deeply disruptive reductions in headcount and lab space due to the broad financial collapse of the telecom industry that began in early 2001. Much of my immediate technical focus was to ensure successful commercial translation

of the TDC technology, which then only existed as prototypes. We worked in close collaboration with a team of engineers in Specialty Fiber Devices (SFD) led by Ben Eggleton, who transitioned from the Lab to SFD around this time. Specifically, we fabricated and provided technical support for devices required for field testing and we set up means for low-volume production in our labs and cleanroom facilities until appropriate infrastructure was installed at SFD.

We also launched new initiatives in unusual microfluidic fiber devices for dynamic gain equalization [44-47] (with Sid Ramachandran, Tom Krupenkine, Shu Yang, Baldwin and others) and ultraminiaturized, high-speed liquid crystal devices for in-line control of the full polarization state of light passing through optical fibers. These latter technologies incorporated microelectrode structures created directly on the cleaved ends of optical fibers, to allow fast, continuous control over the orientation of the director of liquid crystal thin films cast on their surfaces. With custom electronics, optimized drive schemes and specialized liquid crystal materials, we achieved low-loss switching at speeds that set records for liquid crystal devices, exceeding requirements for the most demanding applications in polarization mode dispersion compensation. We successfully demonstrated these devices through tests in 40 Gb/s per channel WDM systems [48-50] (with Bharat Acharya, Ron Pindak, Christi Madsen, Baldwin, Bob MacHarrie and others).

In late 2002, when vast changes were rapidly occurring at the Labs, I accepted a chaired faculty position at the University of Illinois at Urbana/Champaign (UIUC) and the Beckman Institute for Advanced Science and Technology (BI), following a recruitment effort led by Pierre Wiltzius (BI Director at that time) and several other Bell Labs alums on the faculty at UIUC, including Ralph Nuzzo, Paul Braun and Paul Bohn.

I am profoundly grateful to have been given the opportunity to start my independent career at Bell Labs and to collaborate with so many world-class scientists on such a diverse array of exciting topics at the boundaries between science and engineering – including, but not limited to, those described here.

Many measures of accomplishment – Nobel Prizes (10), Turing Prizes (5), multiple revolutionary technologies (transistor, solar cell, CCD, etc) with transformative societal impact -- support the assertion that Bell Labs was the most successful laboratory of all times, perhaps unlikely to be surpassed in the future. This success followed most directly from the co-location of many of the world's most prominent researchers across broad relevant fields of study, working together in a collaborative environment that prioritized major scientific discoveries and impactful engineering innovations over monodisciplinary advances and paper counts. Based on my experiences in the physics laboratory, other notable features of the organization included (1) a long, unmatched history of milestone achievements -- a powerful magnet that attracted the very best young researchers from around the world, (2) a relatively short residence time for individual MTS (~6 years on average, but with a long tail) -- a source of vibrancy from the resulting continuous stream of new hires with different expertise and ideas, (3) a high degree of freedom for MTS to pursue their own ideas as independent principal investigators, typically without rigid timelines, deliverables or significant interference from management -- the basis both for sustained exploratory efforts and for fast responses to unexpected opportunities, (4) a streamlined, versatile funding model to support this mode of operation -- a mechanism that bypassed slow, traditional processes of consensus-based proposal evaluation, (5) a structure that minimized responsibilities outside of those directly related

to research -- a means for MTS to dedicate themselves to their projects without distractions, (6) a collection of well-resourced labs, experienced technicians and exceptional postdoctoral fellows – a way to leverage the unique talents of the MTS, (7) an intellectually challenging culture – an environment that fostered intense, but healthy, levels of competition, (8) a rigorous annual system of performance reviews that rank ordered every MTS within the lab – an assessment of excellence that considered both fundamental (undirected) and applied (directed) research contributions equally and (9) a corporate engineering infrastructure that was closely interfaced to the research enterprise -- an efficient route for converting new scientific knowledge into new technologies.

This distinctive model and signature Bell Labs-ian style left a deep and lasting impression on me. Although I have changed fields of study and home institutions several times since my tenure at the Labs, I have strived to maintain that same powerful approach to research throughout my career, and to share it with collaborators and students alike.

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