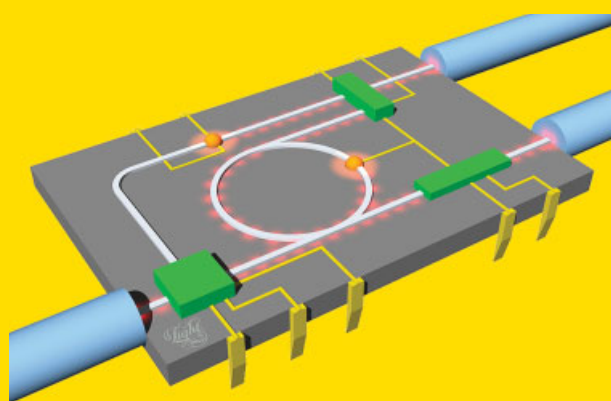


Abstract Techniques for active modulation and control of plasmonic signals in future highly-integrated nanophotonic devices have advanced rapidly in recent years, with recent innovations extending performance into the terahertz frequency and femtojoule-per-bit switching energy domains. As thoughts turn towards the development of practical device structures, key technologies are compared in this review and prospects are assessed for the future development of the field.

Artist's impression of a hybrid opto-/electro-plasmonic chip combining microelectronic and photonic functionalities.



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Active plasmonics: current status

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1. Introduction

“We will only be able to speak about ‘plasmonics’ in the same way we speak about ‘photonics’ when efficient techniques for active manipulation of surface polariton-polariton (SPP) signals are identified.” This challenge was set out in the 2004 paper [1] that coined the term ‘active plasmonic’ - using it to describe a technique for transient switching or modulation of propagating optical-frequency SPP signals. Since that time, a range of material systems, including thermo- and electro-optic media, quantum dots, and photochromic molecules, have been investigated for plasmonic switching applications [2–5]. Most recently, a compact field-effect plasmonic modulator has been demonstrated [6], and the direct optical modulation of SPP propagation on the femtosecond timescale has been reported [7]. In this article we review and compare key experimentally realized active plasmonic technologies: we assess the extent to which the above challenge has been met and consider future challenges and prospects for the field.

The field of ‘plasmonics’, which deals with the unique optical properties of metallic nanostructures, is one of the most fascinating and fast-moving areas of photonics [8–11]. Its explosive growth in recent years has been driven by parallel advances in the capabilities and availability of micro-/nano-fabrication technologies, sample characterization and diagnostic systems, and computational simulation tools, as well as a wealth of potential applications in areas ranging from high sensitivity (bio)chemical sensing [12] to solar power generation [13] and sub-wavelength light-guiding [14]. Particular interest in SPPs - propagating bound oscillations of electrons and light at a metal surface - is based largely on the possibility that they could act as information carriers in future highly-integrated nanophotonic devices. Indeed, plasmonic technologies could transform the chip-scale data transport paradigm by bridging the gaps between current electronic and photonic technologies, combining the nanoscale component dimensions of the former with the speed and bandwidth of the latter and an innate ability to interface with both [15, 16] (Fig. 1).

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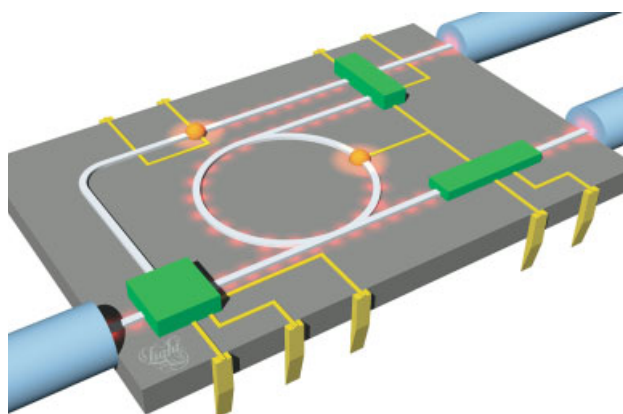


Figure 1 (online color at: www.lpr-journal.org) Artist's impression of a hybrid opto-/electro-plasmonic chip: New and enhanced functionalities are achieved by integrating photonic, electronic and plasmonic systems in single chip-scale devices.

Considerable advances have been made over many years in the development of structures to guide and passively manipulate SPPs [8–10] but it is only in the last few years that attention has turned to the kind of ‘active’ elements that will be required to enable useful plasmonic functionality in device structures.

2. Active plasmonic technologies

The term ‘active plasmonic’ was first used in association with a proposal that high-contrast SPP modulation could be achieved via a light-induced structural phase transition in a nanoscale layer of gallium forming part of a metal/dielectric SPP waveguide [1]. By their very nature, SPP fields are strongly confined at metal surfaces and therefore highly sensitive to refractive index changes within a few tens of nanometres of the surface [17]. The basic idea in active plasmonic systems is to induce such a change in a transient, reversible manner (see Fig. 2). Thus, it was suggested that a transition between the almost ideally-metallic liquid and somewhat semiconductor-like solid phases of gallium in a film just a few tens of nanometres thick at an interface with silica could provide high-contrast modulation of SPPs over a section of waveguide just a few microns long. The plasmonic contrast between gallium’s solid and liquid forms was subsequently demonstrated in an Otto configuration experiment [18] (Fig. 3a) at a free-space light wavelength of 780 nm¹: In a system tuned for coupling to an SPP on a solid gallium surface, a transition to the liquid state, initiated by nanosecond pump pulses at 1064 nm with a fluence of 15 mJ/cm², shifted the system away from resonance and increased the output 780 nm signal power by a factor of 9.4 (9.7 dB); Response times were not resolved in this study; relaxation times as short as 20 ns were recorded.

¹ Free-space light wavelengths, as opposed to SPP wavelength, are used throughout this article.

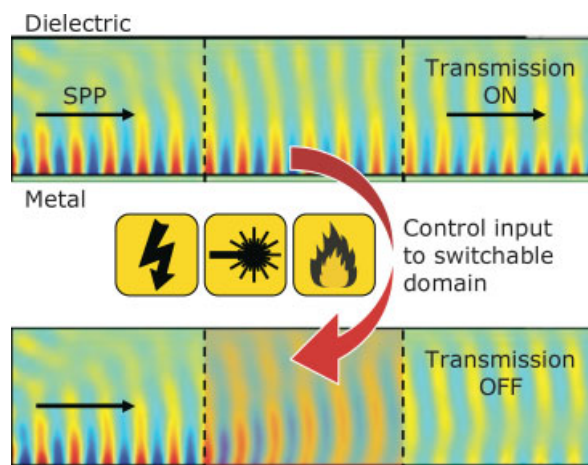


Figure 2 (online color at: www.lpr-journal.org) Generic active plasmonic concept: A transient change in the refractive index of either the metal or dielectric component of an SPP waveguide, induced by an external control excitation (optical, thermal, electronic, etc.), can switch or modulate the intensity and/or phase of a propagating SPP.

Shortly after this, ‘thermo-plasmonic’ devices based on refractive index changes in a thermo-optic polymer (the dielectric component of a long-range SPP waveguide for a wavelength of 1.55 μm) were demonstrated [2] (Fig. 3b). Here, sections of the metal waveguide component – gold strips embedded in the polymer – also acted as the electrical heating elements via which changes in the polymer could be initiated. Mach–Zender interferometric modulator (MZIM) and directional-coupler switch (DCS) configurations were reported: In the MZIM, 8 mW of electrical input power over an active waveguide length of 5.7 mm gave an optical throughput extinction ratio of >35 dB with a response time of ~ 0.7 ms; The DCS achieved a 34 dB extinction ratio at 82 mW with similar response dynamics.

In 2007 a modulator based on SPP-induced generation of excitons in CdSe quantum dots was reported [3] (Fig. 3c): In their ground state, dots coating the surface of a silver waveguide have little effect on the propagation of a signal SPP at a wavelength of 1.426 μm . However, when excited by a co-propagating pump SPP at 514.5 nm they become absorbing at the probe wavelength and attenuate the probe SPP. A change of $\sim 12\%$ (0.55 dB) was observed in the optical signal throughput of a device with an SPP waveguide length of 3.6 μm for an optical pump intensity of ~ 600 Wcm⁻²; This was maintained at modulation frequencies up to 25 MHz (the limit of the detection apparatus), indicating a minimum cycle time of <40 ns (ultimately limited only by the quantum dot exciton recombination lifetime).

The following year, a non-volatile plasmonic switch employing photochromic molecules as the active component was demonstrated [4] (Fig. 3d): The molecules were contained within the polymer component of an aluminium/PMMA waveguide for a signal SPP at 633 nm; They were switched between stable absorbing and transpar-

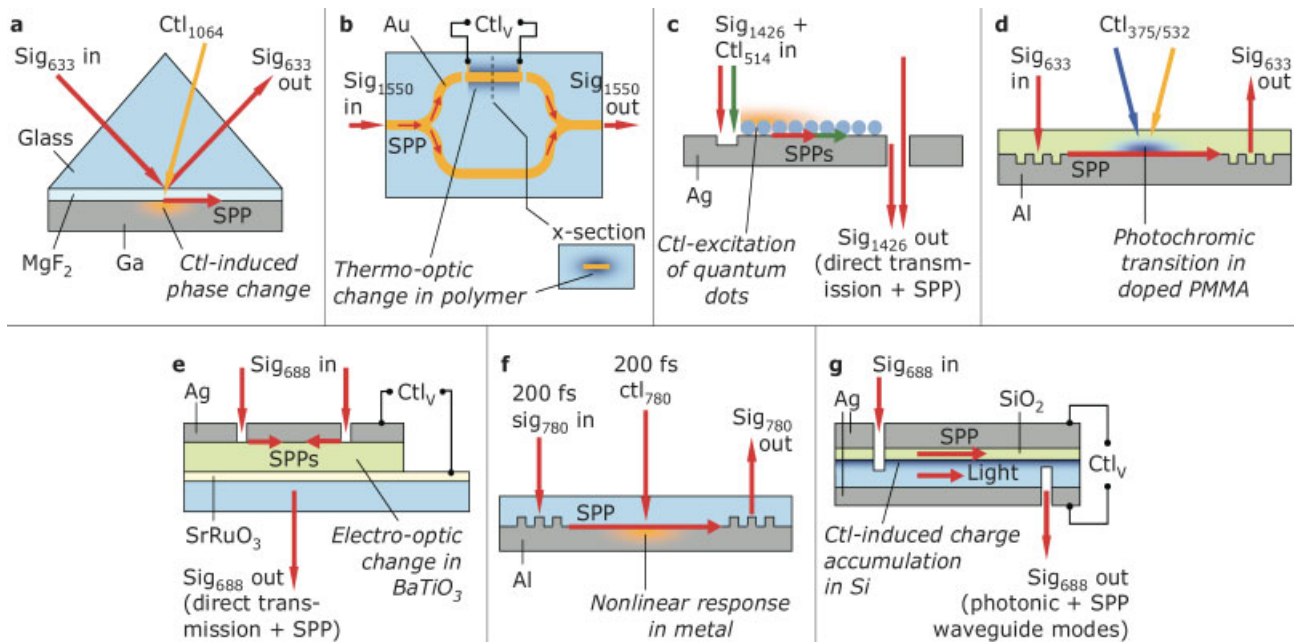


Figure 3 (online color at: www.lpr-journal.org) Schematic configurations of experimental active plasmonic modulators based on: a) a light-induced solid-liquid phase transition in gallium; b) an electrically-driven refractive index change in a thermo-optic polymer; c) absorption activation in CdSe quantum dots; d) photochromic switching in a PMMA matrix; e) electro-optic domain switching and refractive index modification in barium titanate; f) femtosecond optical excitation of aluminium; g) modification of charge distribution in silicon. ‘Sig’ and ‘Ctl’ indicate signal and control inputs respectively with subscripts denoting light wavelength or electrical bias (subscript ‘V’).

ent states using free-space optical pump beams at intensities of $\sim 6 \text{ mW/cm}^2$ (375 nm to switch to the absorbing state, 532 nm to recover the transparent state) and delivered a 6 dB change in the optical output power for an SPP propagation distance of $8 \mu\text{m}$, albeit with very slow transition times of $\sim 10 \text{ s}$.

3. Recent progress

In October 2008, an ‘electroplasmonic’ modulator was reported [5] (Fig. 3e): Here the application of a bias (up to 45 V) across a silver/barium-titanate SPP waveguide induces domain switching in the ferroelectric BaTiO_2 thin film and modifies its refractive index; As part of an interferometric device structure with a length of between 0.5 and $5.3 \mu\text{m}$ such switching modulates the optical output at 688 nm by up to 15% ($\sim 0.7 \text{ dB}$); No data is presented on the temporal dynamics of the effect.

In December 2008, the ultrafast modulation of a femtosecond SPP signal via the direct optical excitation of the metal component of an aluminium/silica waveguide was demonstrated [7] (Fig. 3f): Wavelength-degenerate pump-probe experiments at 780 nm (and a subsequent study covering the range from 765 to 810 nm [19]) reveal a nonlinear interaction between probe SPP and pump light that takes place in the skin layer of the metal; It produces up to 35% ($\sim 1.3 \text{ dB}$) modulation of the probe for a pump fluence of

13 mJ.cm^{-2} over an SPP propagation distance of $5 \mu\text{m}$; The response shows two components, a ‘fast’ component with transition times shorter than the 200 fs pulse duration used in the experiments, and a ‘slow’ component (emerging at higher pump fluences) with a decay constant of $\sim 60 \text{ ps}$; The fast component is sensitive to the mutual orientation of the pump polarization direction and the electron oscillation direction in the SPP - only appearing when they are parallel; Both depend strongly on the proximity of the excitation wavelength to aluminium’s interband absorption peak at $\sim 1.5 \text{ eV}$.

Finally, in February 2009, a metal-oxide-semiconductor (MOS) field-effect plasmonic modulator operating at $1.55 \mu\text{m}$ – a ‘plasMOSstor’ – was reported [6] (Fig. 3g): The device employs a four-layer metal-MOS-metal (Ag- SiO_2 -Si-Ag) waveguide structure that supports both photonic and plasmonic modes, and has a transmission coefficient determined by interference between the two; Application of a bias of 0.75 V drives the MOS into accumulation, changing the Si index and cutting off the photonic mode, and thereby changing the transmission; A peak modulation depth of 4.56 dB is observed in a device $2.6 \mu\text{m}$ long; Switching times are found to be shorter than the 10 ns limit of the experimental apparatus. Indeed, it is suggested that the fundamental limit on switching speed is that of the formation of the MOS accumulation layer (as it is in a conventional small-geometry MOS field-effect transistor), and therefore that the plasmostor could operate at gigahertz frequencies.

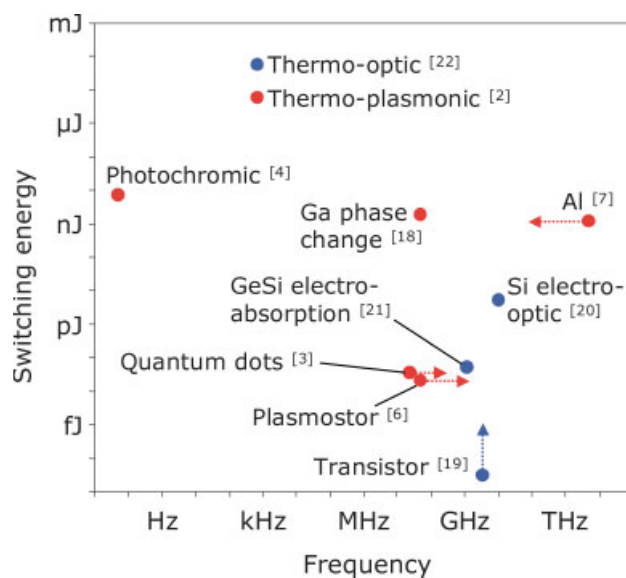


Figure 4 (online color at: www.lpr-journal.org) Active plasmonic switching performance: Switching energy and cycling frequency characteristics for active plasmonic modulation technologies [red] (where necessary an interaction length of $8\ \mu\text{m}$ within a waveguide $1.5\ \mu\text{m}$ wide has been assumed to estimate switching energy) and a selection of existing or competing electronic/photonics technologies [blue]. Dashed arrows indicate: possible higher frequency operation of quantum dot and plasmistor devices (instrumentation-limited measurements); an operating frequency limited by the slow response component in the Al-based device at high pump fluence; and an energy/bit limit for electronic data processing set by interconnect rather than transistor performance.

4. Comparative performance

Fig. 4 shows a switching energy vs. modulation frequency map for the active plasmonic technologies outlined above. These are only two of the numerous criteria (e.g. switching contrast, size, long-term stability, ease of fabrication, compatibility with existing electronic/photonics technologies, etc.) on which they might be evaluated but provide a good basis for comparative discussion at this early stage in the development active plasmonic systems. Also highlighted on this diagram are characteristics for a microprocessor transistor [20] (data are presented for a single transistor, though it should be noted that in device structures the energy/bit limit is increasingly set at a somewhat higher level by the interconnects between components [15, 16]), and electro- [21, 22] and thermo-optic [23] photonic modulators – if plasmonic technologies are going to augment and ultimately supplant such devices then they should at least match their performance.

For high-speed data processing, the thermo-plasmonic and photochromic devices reported to date are too slow. Which is not to say they will not find valuable application elsewhere: The thermo-plasmonic system [2] provides high switching contrast in a configuration compatible with basic

planar fabrication techniques and, by virtue of inherently positioning the control electrode at the maximum of the SPP mode field, is considerably more efficient than conventional thermo-optic modulators [23]; Though susceptible to photo-degradation, the photochromic system [4] provides good intrinsic modulation contrast (as opposed to a level enhanced in an interferometric arrangement) and is uniquely bistable among the switchable media described here so far – it can remain in either state indefinitely without drawing power and may therefore be suited to memory applications. Other switchable molecules may offer the possibility of much faster transient signal modulation. J-aggregates, for example, have been used to control the plasmon-mediated extraordinary optical transmission of a nano-hole array on the picosecond timescale [24] but have not yet been tested in application to the control of a guided SPP signal.

The gallium-based modulator [1, 18] also provides good intrinsic contrast and gets closer to the desired operating frequency but is still limited by a thermal relaxation process taking at least a few tens of nanoseconds. The involvement of a liquid phase in the device is less problematic than one might imagine: the transition to liquid occurs only in a nanolayer of material at a solid dielectric interface and is stable against repeated cycling [25]. The main obstacle to the use of gallium is its intermetallic chemistry: for want of a better description it is ‘corrosive’ to metals like silver, gold and aluminium which are favoured in SPP waveguides – it aggressively penetrates grain boundaries and forms intermetallics. Alternative ‘phase-change media’, such as transition metal oxides [26] and the chalcogenide glasses on which today’s optical data storage technologies and next-generation electronic memories are based [27], may be better suited to commercial application. The chalcogenides in particular promise fast, high-contrast, non-volatile, optically- and electrically-driven switching functionality [28].

The quantum dot modulator [3] offers a modest contrast ratio (in an interferometric configuration), an operating frequency limit at least as fast as the gallium phase-change device, and a particularly low switching energy. Most notably, this configuration allows for, or rather relies on ‘all plasmonic’ modulation: one SPP controlling another.

The ultrafast nonlinearity of aluminium provides a plasmonic modulation mechanism several orders of magnitude faster than any other in a simple planar waveguide architecture that does not require the introduction of any dedicated switching media [7, 19]. The modulation depth is good (and may be improved with judicious waveguide design) and the required switching energy is relatively high. The terahertz operating frequencies offered by this technique may in fact be considered excessive for next-generation plasmonic data-processing applications [15, 16] but the experimental result is indicative of a growing interest in ultrafast and nonlinear plasmonics [29–31]. It invites further investigation of what can ultimately be achieved in these areas and such studies will inform the future development of technologically relevant device structures and operational modes for plasmonics in the same way that studies of ultrafast

and nonlinear optical phenomena have contributed to the advancement of photonic technologies.

The plasmistor [6] comes closest to the few-gigahertz operating frequencies of today's microelectronic data processors. The reported interferometric configuration achieves reasonable modulation contrast and does so at very low switching energies (lower than those required in photonic modulators [21, 22]), though the signal propagation losses are relatively high. This article illustrates an emerging trend in active plasmonics towards consideration of practical device structures. Indeed, it proposes and models a chip-scale optically driven modulator that could be manufactured using current SOI (silicon-on-insulator) fabrication techniques.

5. Outlook

In answer to the call for active SPP modulation techniques, a range of technologies have been developed, each with their own advantages and disadvantages. Alternative switchable media (transition metal oxides [26], chalcogenides [27, 28], J-aggregates [24], etc.) may yet bring new or improved functionalities but with recent experimental devices already pushing performance limits into the terahertz modulation frequency and femtojoule/bit switching energy domains, the challenge now is to develop device structures that are efficient, compact and compatible with existing CMOS and/or SOI fabrication processes. Such plasmonic components could, in the first instance, simply bring photonic and electronic functionalities together on a single chip to create powerful hybrid devices that take advantage of the two technologies' respective strengths.

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