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Review

Magneto-Optics Effects: New Trends and Future Prospects for Technological Developments

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Abstract: Magneto-optics (MO) is an effervescent research field, with a wide range of potential industrial applications including sensing, theranostics, pharmaceutics, magnetometry, and spectroscopy, among others. This review discusses the historical development, from the discovery of MO effects up to the most recent application trends. In addition to the consolidated fields of magnetoplasmonic sensing and modulation of optical signals, we describe novel MO materials, phenomena, and applications. We also identified the emerging field of all-dielectric magnetophotonics, which hold promise to overcome dissipation from metallic inclusions in plasmonic nanostructures. Moreover, we identified some challenges, such as the need to merge magneto-chiroptical effects with microfluidics technology, for chiral sensing and enantioseparation of drugs in the pharmaceutical industry. Other potential industrial applications are discussed in light of recent research achievements in the available literature.

Keywords: magneto-optics; magneto-plasmonics; magnetophotonics

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1. Introduction

Magneto-optics refers to changes in the properties of light when it is transmitted or reflected in the presence of a magnetic field (externally applied or from a magnetized material medium). These phenomena were first discovered by Michael Faraday in 1845 [1]. He noticed that a linearly polarized light beam undergoes a polarization rotation when it propagates parallel to an externally applied magnetic field. Just over 30 years later, the Reverend John Kerr observed the corresponding effect when he studied the reflection of linearly polarized light from the surface of a magnet [2,3]. These magneto-optics (MO) effects, conventionally called Faraday and Kerr MO effects (in transmission and reflection, respectively), not only helped establish the theory of electromagnetism in the late 19th century, but also laid the foundation for several technological achievements. The Faraday effect, for example, has the unique property of breaking time-reversal symmetry and Lorentz's reciprocity theorem [4], which has been utilized for the development of non-reciprocal optical devices and laser systems [5,6]. The Kerr effect, on the other hand, allows a way to measure or visualize the magnetic state of material media, and has therefore been used for spectroscopy [7–10] and data-storage applications [11–13]. More recently, the advent of nanotechnology and modern nanofabrication tools motivated the exploitation of MO phenomena at micro- and nano-scale levels for a new era of devices, with their optical properties actively manipulated by applied magnetic fields. However, the latter is challenged by weak MO effects at visible and infrared wavelengths [14].

The search for enhanced MO effects at the nanoscale has been led (almost exclusively) by plasmonics over the last few decades, in what is commonly known as magnetoplasmon-

ics [15]. This last approach employs the localized and strongly enhanced plasmonic fields to improve the MO activity of nanostructures [16,17]. Plasmonics refers to the resonant coupling of light with the collective oscillations of the electron cloud (called plasmons) on a metallic surface [18], with the unique ability to concentrate light into deep-subwavelength volumes (enabling devices with unprecedented miniaturization levels) [19]. Magnetoplasmonics has therefore been realized with nanostructures made of pure noble metals [16], pure ferromagnetic metals [20–22], hybrid noble-ferromagnetic metals [23], and hybrid noble metal-ferromagnetic dielectrics [24,25], with applications in (bio)sensing [26–29], nonreciprocal waveguides [30] and magnetic control of the optical chirality [31], among others. Despite these advances, magnetoplasmonic applications are still hampered by the intrinsic losses of metallic inclusions, which has motivated the search for new materials and structures to overcome these challenges. In particular, the reduced level of losses from dielectric ferromagnetic materials [32–35], combined with their integrability with silicon photonics [36,37], has motivated a lot of recent research [38–43]. Moreover, the design and development of hyperbolic MO metamaterials has also gained interest during the last few years [44–48].

In this review paper, we advocate the importance of MO effects for future active nanophotonic devices and their perspectives for industrial applications. The microscopical and macroscopical theory of MO effects (including magnetoplasmonics) have been discussed in several previous works [49–54], to which we refer interested readers. To avoid repetition, we focus our attention here to discuss the most recent trends and future prospects in magnetophotonics, i.e., magneto-optic nanostructures, exemplifying the merging of MO with surface enhanced Raman spectroscopy (SERS), which has already been applied for sensing and magnetic separation of biological moieties.

2. A Brief Review on Magnetoplasmonics

Due to the importance of nanotechnology and magnetophotonics, and also to provide the reader with a historical context, we prefer to start with a brief history of plasmonics. The first experimental evidence of surface plasmon resonance (SPR) phenomenon was observed by Wood in 1902 [55] while measuring the reflectance spectra of a metallic grating. In particular, Wood observed a sharp discontinuous change in the reflectance (around a specific frequency), which was called the Wood's anomaly. Then, in 1907, Rayleight discovered that the frequencies at which anomalies occur are intimately related to the incident angle and wavelength (as well as to the angle of emergence of the diffracted waves), providing a qualitative explanation for the Wood's anomaly [56,57]. However, it was not until 1941 that Fano demonstrated that the physical principle behind the Wood's anomaly stems from the excitation of surface waves at the metallic/dielectric interface [58]. Indeed, Fano indicated the similarity between the Wood's anomaly in the grating and the surface waves in the attenuated total reflection (ATR) effect. Furthermore, in the early 1950s, Pines and Bohm [59–62] showed in a series of papers that conduction electrons in a metal (considered as a free electron gas) can describe collective oscillations, whose quantizations were called plasmons. Based on these previous achievements, Ritchie derived the dispersion relation of SPRs in 1957 [63], which was experimentally demonstrated two years later by Powell and Swan [64]. Merging the ATR analogy (described by Fano) with the SPR dispersion relation, Kretschmann [65] and Otto [66] proposed the use of prism couplers (high-refractive-index incident media) for the excitation of SPRs in flat planar metallic surfaces. On the other hand, the time harmonic oscillation of the electric field component of light forces the density of free charges in metallic nanoparticles to oscillate between upper and lower boundaries. This oscillation exhibits a maximum amplitude at the resonant frequency, analogous to a forced harmonic oscillator [67]. Although in 1904 Maxwell–Garnett used an effective dielectric permittivity theory to explain the colors of glasses containing small metallic nanoparticles [68], the full electromagnetic theory of light scattering and absorption by colloidal metallic nanoparticles was only developed in 1908 by Mie [69]. Plasmonic resonances in metallic nanoparticles are radiative localized (non-

propagating) SPRs (LSPRs), contrary to SPRs in gratings or flat planar metallic surfaces, which are surface-guided modes achieved under phase-matching conditions. These basic plasmonic platforms are illustrated in Figure 1a,b, where the order of electromagnetic decay lengths is also illustrated for each case.
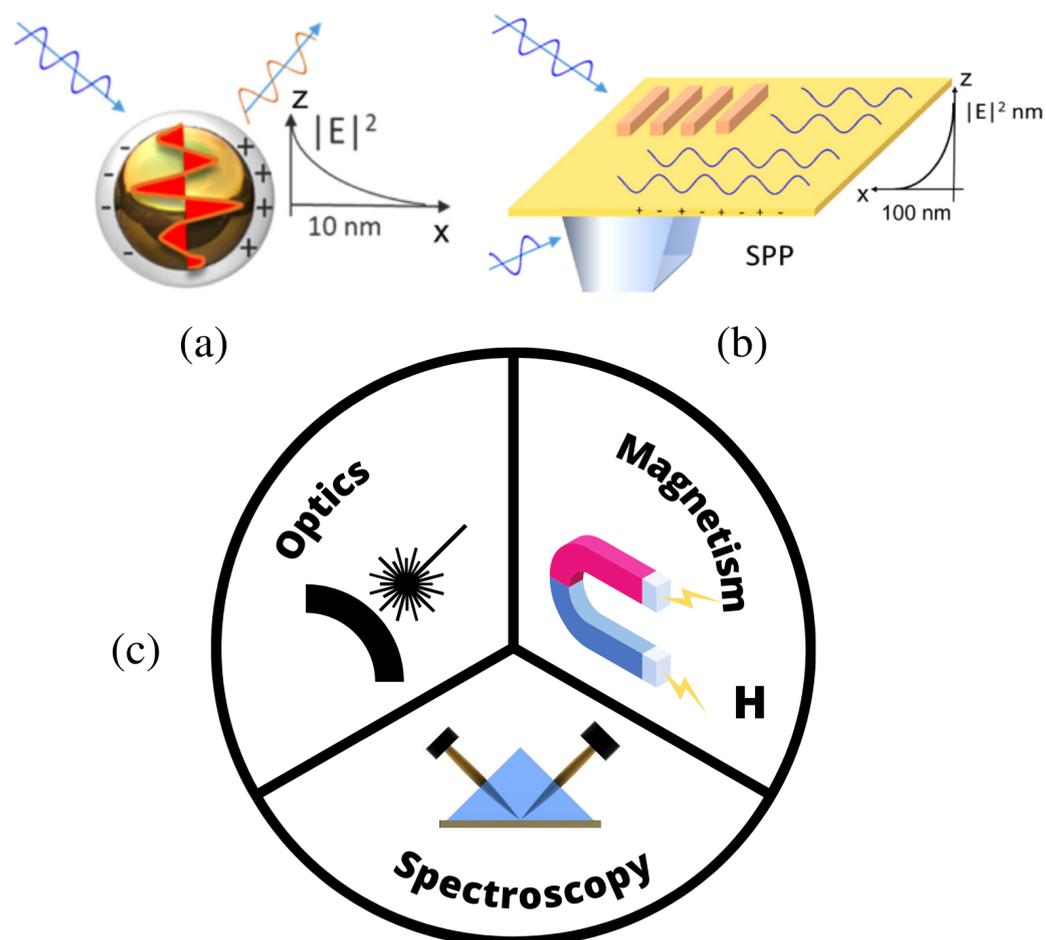

Figure 1. (**a**) Localized surface plasmon resonances (SPRs) and (**b**) propagating SPRs. (**c**) Magnetoplasmonics: merging of optics, magnetism and spectroscopy. Figures (**a**,**b**) were adapted from the American Institute of Physics, 2021 [70]. Copyright 2021 American Institute of Physics.

Since the first experimental demonstrations of SPR applications in gas sensing and biosensing, made four decades ago by Nylander and Liedberg et al. [71,72], there has been intense research activity to enable this technology for the detection of small analytes or in very dilute solutions [73]. However, due to very small changes in the refractive index (RI) of the analyte medium, the resolution of SPR (bio)sensing devices is largely limited by the overlap between nearby resonances. In the search for alternatives to surpass this last drawback, magnetoplasmonic biosensing, i.e., merging plasmonics with magnetism and spectroscopical techniques (illustrated in Figure 1c), has emerged as a promising alternative. The first experimental demonstration of magnetoplasmonic sensing was made by Guo et al. [74], in 1999, who used MO modulation to monitor the phase shift resulting from the minute change of the angle of incidence, demonstrating improved sensing resolutions compared to the conventional SPR approach. Then, in 2006, Sepúlveda et al. [26] demonstrated resolution improvements of up to three orders of magnitude in biosensing applications when using the sharp curves from plasmonically enhanced transverse MO Kerr effect (TMOKE) instead of broad reflectance SPR lines. A prototypical MOSPR experimental setup for sensing is shown in Figure 2a, where a magneto-plasmonic waveguide combining a noble metal (Au in this case) and a ferromagnetic metal (Co in this case) is used in the

Kretschmann-like configuration for improved sensing performance (resolution, sensitivity and detection limit). The MOSPR sensor in Figure 2a is composed by a Au (5 nm)/Co (3 nm)/Au (21 nm) tri-layer film on glass substrate and covered by air (the superstrate). A prism coupler of glass is also used for the ATR mechanism. The reflectance (R) associated with the demagnetized system ($\mathbf{M} = \mathbf{0}$), represented with solid blue circles (see the left axis), is comparatively plotted with the corresponding TMOKE $= \frac{\Delta R}{R} = \frac{R(+M)-R(-M)}{R(+M)+R(-M)}$ values (represented by solid orange circles in relation to the right axis) around a minima of R. From this last result it can be clearly seen that TMOKE curves are sharper than SPR ones, which is used to improve the quality factor and the sensing performance of the structure, as previously mentioned. The superstrate is then successively injected with concentrations of 1%, 2%, 3%, and 4% of ethanol gas diluted with nitrogen, with the corresponding time transient measurements in Figure 2c,d, respectively. As noticed, the signal-to-noise ratio is better for $\Delta R/R$ than that for reflectivity R, showing improved sensitivity by using the MO-SPR sensing. It is worth mentioning that further resolution improvements can be achieved through combined advanced modeling and nanopatterning methods [75–79]. Nevertheless, the MOSPR sensing approach may be inappropriate for detection of very high analyte concentrations, as can be seen in Figure 2d. This last behavior is explained through the close relationship of the optimized TMOKE amplitudes with the SPR phase-matching condition, considering that the latter is lost when the changes in the refractive index are too high.
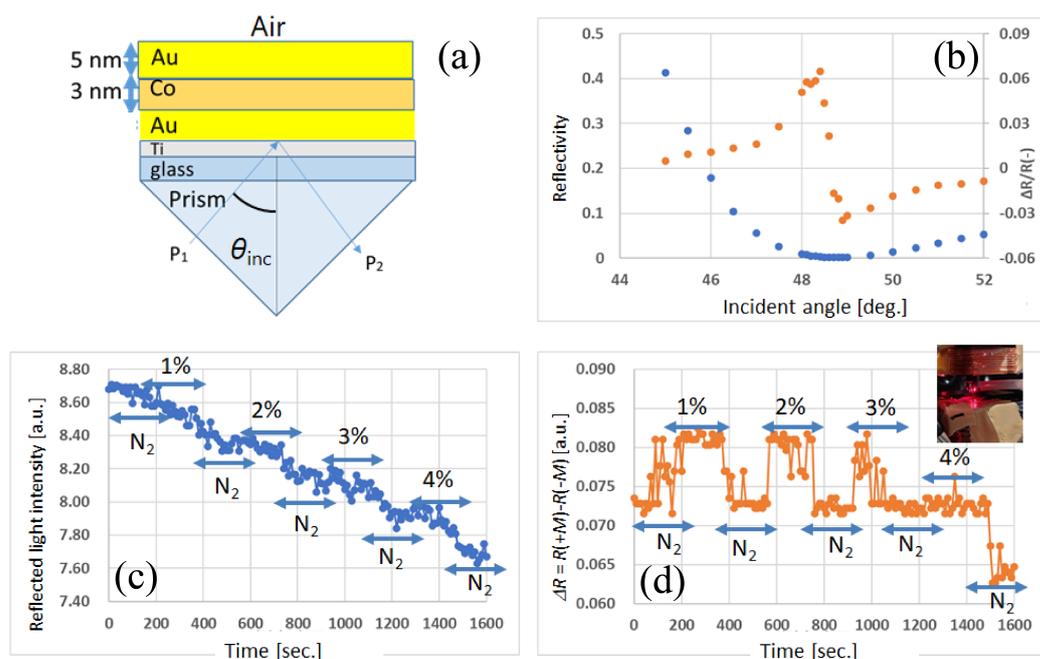

Figure 2. (**a**) A schematic diagram of the fabricated Au/Co/Au MOSPR sensor. A quartz glass prism is mounted on top of the glass substrate, and the light is coupled with an incident angle θ_{inc}. (**b**) The reflectivity measured in air without magnetic field (left axis), and TMOKE $= \Delta R/R$ (right axis). Time transient of (**c**) the reflected light intensity and (**d**) TMOKE upon ethanol gas injection at concentrations of 1, 2, 3, and 4%. In the inset is a photo of the MOSPR sensor chip that was mounted on a quartz prism and positioned between a pair of coils. This chip was part of the Au/Co/Au construction.

Achievements in SPR magnetoplasmonics are not limited to (bio)sensing. For example, the plasmonically enhanced magneto-optic effect is also being actively exploited for new integrated active nanophotonic devices. In particular, high-speed non-reciprocal magnetoplasmonic waveguides have been actively developed during the last years [80–82]. These latter devices enable unidirectional light propagation by actively manipulating the insertion losses for forward and backward modes in a magnetoplasmonic waveguide. This

active tuning is achieved by using an external magnetic field to manipulate the magnetization state of a building ferromagnetic material in the waveguide, as depicted in Figure 3, which in turn alters the insertion loss level in the waveguide. An important application of non-reciprocal waveguides is the stable operation of semiconductor lasers, preventing them from unwanted backward reflection in the optical transmission path. Instead of free space optical isolators consisting of Faraday rotators (magnetic garnet) and two polarizers, semiconductor optical isolators based on the non-reciprocal loss, have been studied, where ferromagnetic metal thin films are deposited at a part of the waveguides. Transversely magnetized ferromagnetic metal provides different propagation loss depending on the magnetization direction or propagation direction, owing to the time-inversion asymmetry. Significantly, on-chip optical isolators can be realized through monolithic integration of semiconductor optical isolators with semiconductor lasers, simplifying the system and avoiding the work of physical alignment between the lasers and isolators [83]. In Figure 4a, we show a cross-section scanning electron microscopy (SEM) micrograph of a semiconductor optical isolator, comprising a ferromagnetic Co thin film on an InP substrate. The vertically magnetized Co thin film provides optical isolation for transverse electric (TE) mode light by using the TMOKE. The experimental setup for optical isolation measurements is shown in Figure 4b, and results showing optical isolation of up to 45 dB/mm are presented in Figure 4c.

The magnetic control of chiroptical activity is a new and flourishing application of magnetoplasmonics that is increasingly gaining attention [84–89]. The term chirality, used for the first time in 1894 by Lord Kelvin [90], refers to objects that are non-superimposable on their mirror images. Our hands are, perhaps, the most universal example. Because of this analogy, both enantiomorphs (from the Greek's composition enantios = opposite + morphe = form) are conventionally classified as L- (left) and R-enantiomorphs (right) or, when referring to molecules, L- and R-enantiomers. In contrast to achiral (without chirality) environments, where both enantiomers of a chiral molecule (sharing the same stoichiometric molecular formula) exhibit the same physical and chemical properties, molecular chirality plays a key role in the biochemical and biological activity of molecules. In fact, because the elementary building blocks of living organisms (e.g., amino acids and sugars) are chiral, biochemical reactions are inherently chiral [91,92]. The latter is crucially important in pharmacology, where therapeutic effects are associated with a single enantiomer and the other is ineffective or induces serious side effects [93–95], as was demonstrated by the thalidomine disaster in the late 1950s [96–98].
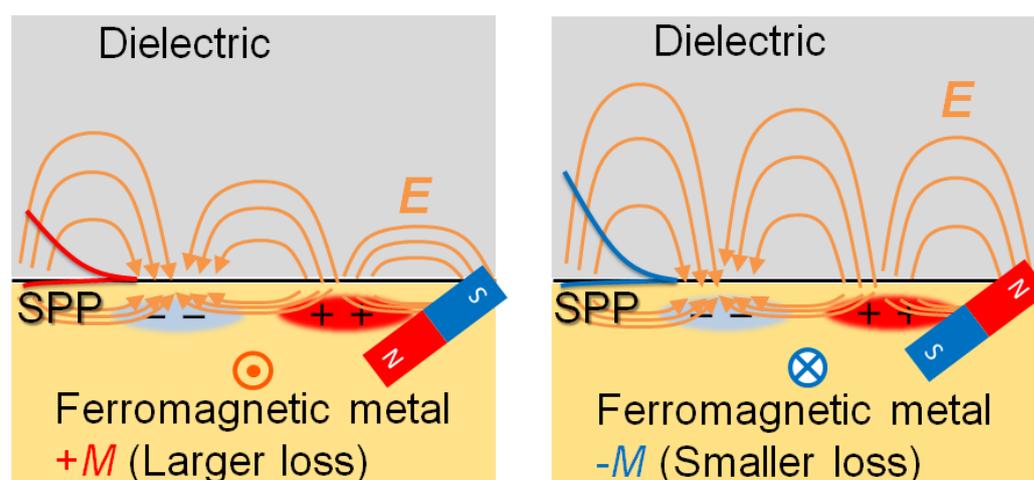

Figure 3. Illustrative representation for the active tuning of insertion losses in a magnetoplasmonic waveguide.
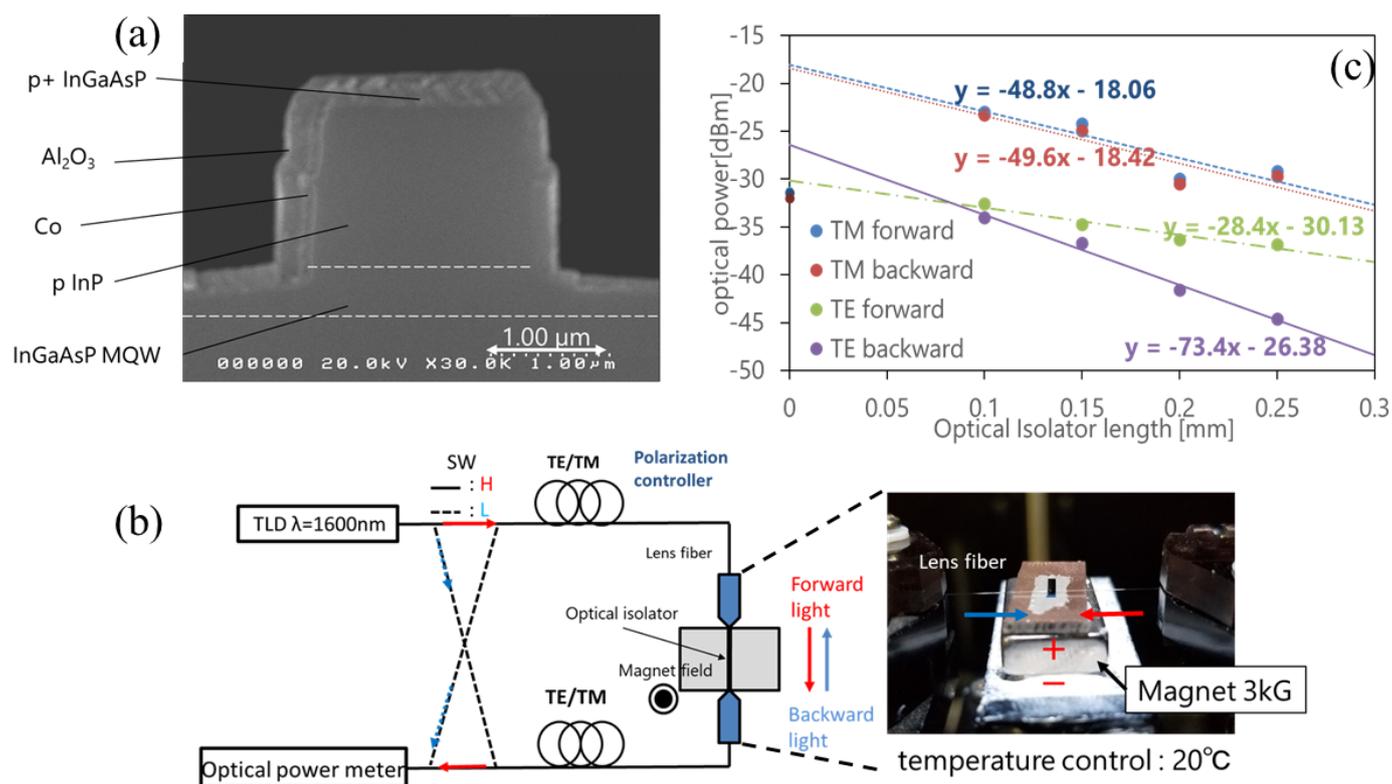
Figure 4. (**a**) A cross-sectional SEM image of the TE mode semiconductor optical isolator. (**b**) A schematic diagram of the experimental setup for measuring the characteristics of semiconductor optical isolators. (**c**) Forward and backward propagation loss as a function of the length of the semiconductor optical isolators.

Although physical and chemical techniques (e.g., circular dichroism and chromatography) are currently used for recognition and separation of enantiomers in the pharmaceutical industry, their performance is still limited to high concentrations or large analyte sizes [99,100]. On the other hand, the Faraday rotation effect is itself chiral. In fact, the physics behind magnetic circular dichroism (MCD), i.e., the differential absorption of left and right circularly polarized light, stems from the Faraday effect [53]. Therefore, through the unique sensing capabilities of plasmonics (using chiral or achiral geometrical designs [101]) with an active CD modulation mechanism [102], the combination of the magnetoplasmonic Faraday effect with circular dichroism (CD) spectroscopy (commonly known as magneto-chiroptical effect) may allow improved detection limits with higher resolutions [103,104]. Figure 5a–c illustrates the magneto-chiroptical application in tuning the extrinsic chiroptical activity from a perforated Au slab, comprising an achiral hexagonal arrange of circular nanoholes [31], whereas Figure 5d,e show the circular differential transmission (CDT) from a chiral arrange of Au and Ni nanoparticles [105]. As was recently indicated in ref. [70], these enhanced magneto-chiroptical effects can be simultaneouly used with magnetoplasmonically enhanced optical forces [106,107] for optical enantioseparation of chiral samples in the pharmaceutical industry.
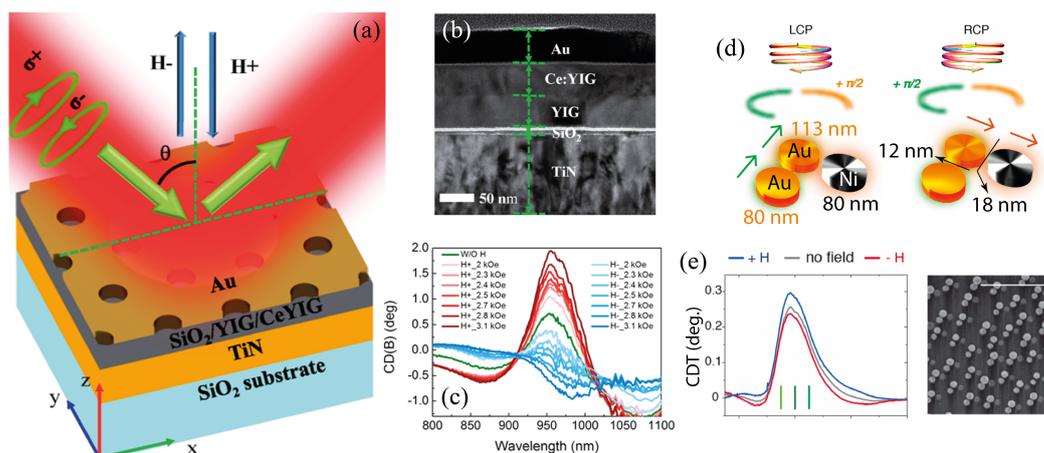
Figure 5. (**a**) Schematic of the magnetoplasmonic multilayer nanostructure for switching the extrinsic optical chirality by using the magneto-chiroptical effect. The applied magnetic field can be alternated between the $+z$ (H+) and $-z$ (H−) directions, as indicated, and the incident CPL can be RCP ($\sigma+$) or LCP ($\sigma-$). (**b**) Different material layer thicknesses are highlighted by using a cross-sectional TEM micrograph. (**c**) Results of magneto-chiroptical effect are comparatively shown with the extrinsic chiroptical activity (without W/O H, see the green line) through CD measurements, for an angle of incidence $\theta = 45°$, by using magnetic field amplitudes varying from -3.1 kOe to $+3.1$ kOe (applied along the z axis, as illustrated). Reprinted with permission from Qin et al., ACS Nano **14**, 2808 (2020) [31]. Copyright 2020 American Chemical Society. (**d**) Magnetoplasmonic chiral arrange of Au and Ni nanoantennas on a substrate. Green and orange arrows are used to indicate longitudinal resonances of bimetallic nanoparticles, excited with quarter-period phase delay. (**e**) Experimental results for the CDT associated to H+ and H- are comparatively shown with the case without H. Reprinted with permission from Zubritskaya et al., Nano Lett. **18**, 302–307 (2018) [105]. Copyright 2018 American Chemical Society.

3. A Brief Review on All-Dielectric Magnetophotonics

In spite of the tremendous technological achievements using magnetoplasmonic nanostructures, the use of metallic building blocks tacitly implies high absorption losses due to the intrinsic Joule effect. Although researchers have tried to improve the structure and material quality of plasmonic systems [108,109], to reduce losses, energy dissipation remains high and unavoidable. The most recent attempts include the design and development of magnetoplasmonic metamaterials [44–48] that, while demonstrating interesting physical phenomena, are still limited by the use of metallic inclusions. On the other hand, all-dielectric nanostructures exhibiting low optical absorption at visible wavelengths and lossless optical response at the infrared range [110] are gaining interest for modern applications. This interest is mainly fueled by their unique set of optical resonant features, including electric, magnetic and anapole resonances [111–113]. In this context, one of the major drawbacks is the mismatch between the lattice parameters of dielectric MO materials and CMOS compatible semiconductors, which has motivated research on novel materials and manufacturing methods [114–116]. For example, cerium substituted yttrium iron garnet (Ce:YIG) films have been grown on gadolinium gallium garnet substrates, with orientations (100), (110) and (111), through the pulsed laser deposition method [116]. Experimental results for the off-diagonal permittivity component (ε_2) and extinction coefficient (k), shown in Figure 6a,b, demonstrate high MO activity with relatively low optical absorption. These results are of special interest for the development of future non-reciprocal and energy-efficient integrated photonic circuits, with applications in photonic computing and data transport architectures. Indeed, there are currently available several new dielectric MO materials that are monolithically integrable with CMOS technology [36,37,40]. Other recent approaches include the development of transparent MO ceramics [117,118], with applications in Faraday isolators for high-power lasers [119,120].
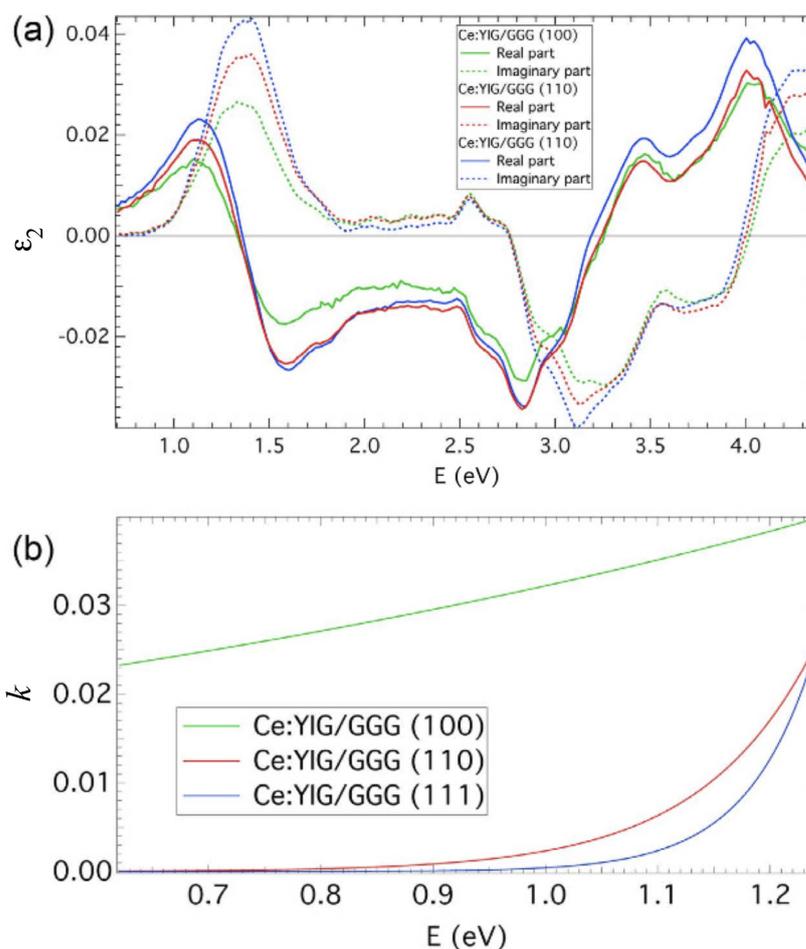
Figure 6. The real and imaginary parts of the off-diagonal permittivity component (ε_2) and the extinction coefficient (k) are shown as a function of the incident wavelength in (**a**,**b**), respectively, for three Ce:YIG samples on GGG substrates. Adapted with permission from ref. [116]. Copyright 2016 Springer Nature.

In addition to new low-loss all-dielectric MO materials, nanostructured all-dielectric MO platforms are also needed to compete with the well-established magnetoplasmonic mechanism for amplified MO effects. Recent achievements include high-Q MO metasurfaces for advanced light control in dual polarizations [33], as schematically represented in Figure 7a. This metasurface consists of a two-dimensional array of Bi-substituted iron-garnet nanopillars, fabricated by electron-beam lithography, on an ultrathin Bi-substituted iron-garnet slab (epitaxially grown on a gadolinium gallium garnet (GGG) substrate). A SEM micrograph of the fabricated structure is shown in Figure 7b. The diffraction properties of the nanopillar lattice enables the coupling between the incident light and the quasi-waveguide modes in the metasurface. Significantly, the nanopillar array exhibits resonances for both TE and TM (transverse magnetic) polarizations, whose field profiles are shown in Figure 8a–d for $TE_{0,1}$ and $TM_{1,0}$, allowing active manipulation of the propagation through changes of the in-plane magnetization (**M**, illustrated in Figure 7a). Other approaches use metal-free MO hyperbolic metamaterials, which can be fabricated with oxide-nitride nanostructures [46]. All these advances have an impact on the photonic integrated circuits (PICs) industry for dynamic optical signal processing at the chip scale [41] and for on-chip MO memories [121]. Applications also include (bio)sensing [122,123], ultrafast optical spin dynamics manipulation [124], and light filtering and modulation [125].
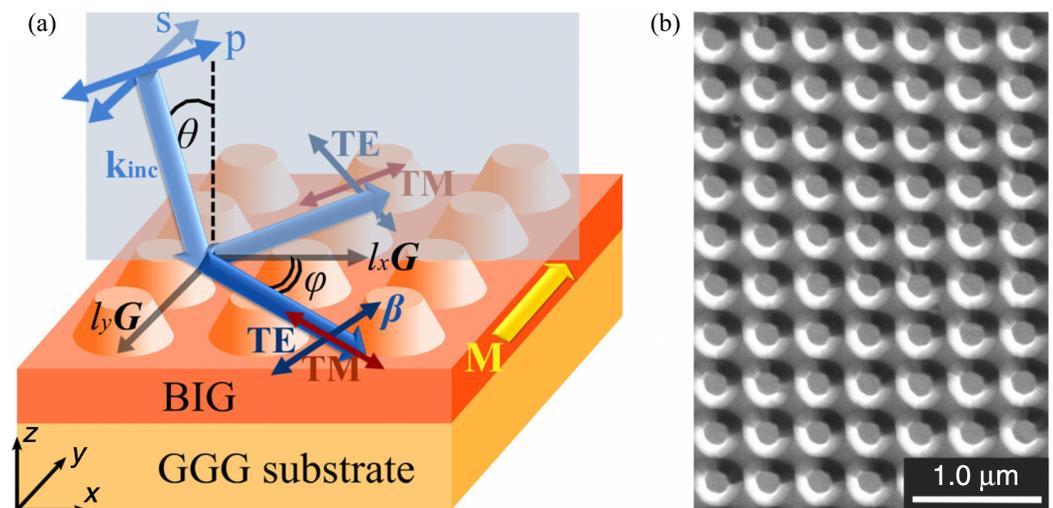
Figure 7. (**a**) Schematic of the metasurface and mode excitation. (**b**) SEM picture of the fabricated microstructure. Adapted with permission from ref. [33]. Copyright 2020 Springer Nature.
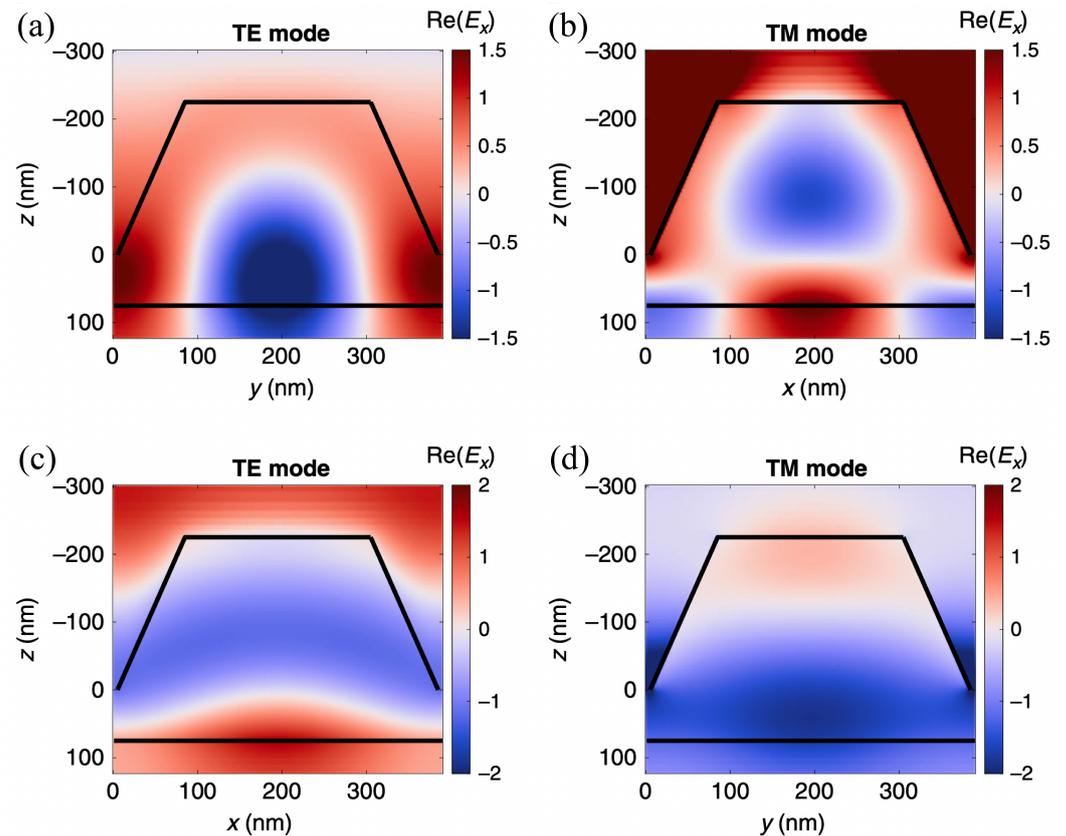

Figure 8. Real part of the electric field component E_x associated with the resonant electromagnetic field distributions for the (**a**,**c**) $TE_{0,1}$ (propagating along the y-axis) and (**b**,**d**) $TM_{1,0}$ (propagating along the x-axis) modes. Numerical results, shown inside one period of the metasurface (the cross-sections are taken at the center of the nanopillar), were calculated for a TM incident wave under normal incidence. Adapted with permission from ref. [33]. Copyright 2020 Springer Nature.

4. A Timeline View of Magneto-Optics and Its Applications

So far, we have discussed MO from their discovery up to the fusion with nanotechnology for advanced applications. Before providing our perspective on integrated magneto-photonics and its future potential industrial applications, we want to make a timeline of MO effects. Figure 9 illustrates the successive developments that have had strong influence

on the development of MO-based technology. In particular, the field of MO was relatively dormant until the mid-20th century, when novel light sources and laser technology became available [126,127]. Then, the advent of nanotechnology and modern simulation methods prompted the study of MO effects at micro- and nano-scale [128–131]. Indeed, merging femtosecond lasers with nanostructured surfaces has been recently used for ultrafast all-optical modulation of MO effects [132–134]. These current advances have made possible the emergence of time-resolved magneto-optical spectroscopy, which, in turn, allows the investigation of phenomena such as the spin relaxation of non-equilibrium photo-excited carriers, transient modifications of the ferromagnetic order, and the photo-induced dynamics phase transitions, to name just a few. In addition, the strongly enhanced MO effects in nanodevices (induced by light–matter interaction at the nanoscale) are revolutionizing several fields, including medicine [135,136], radiative energy transfer [137–139], and electronics [140,141]. On the other hand, recent studies are opening new possibilities to further understand magnetic and MO effects and their interaction with other physical quantities. For example, there are works demonstrating magnetization switching through the use of elastic strain pulses [142]. Furthermore, new microscopical theories to explain MO phenomena, based on the quantization of topological MO effects in antiferromagnetic materials [143], are promising to replace the conventional microscopic theory. Moreover, the development of new sensitivity metrics has made it possible for users to compare the performance of different kinds of sensors in a manner that is direct and independent of the biomolecular interaction between the sensors [144].
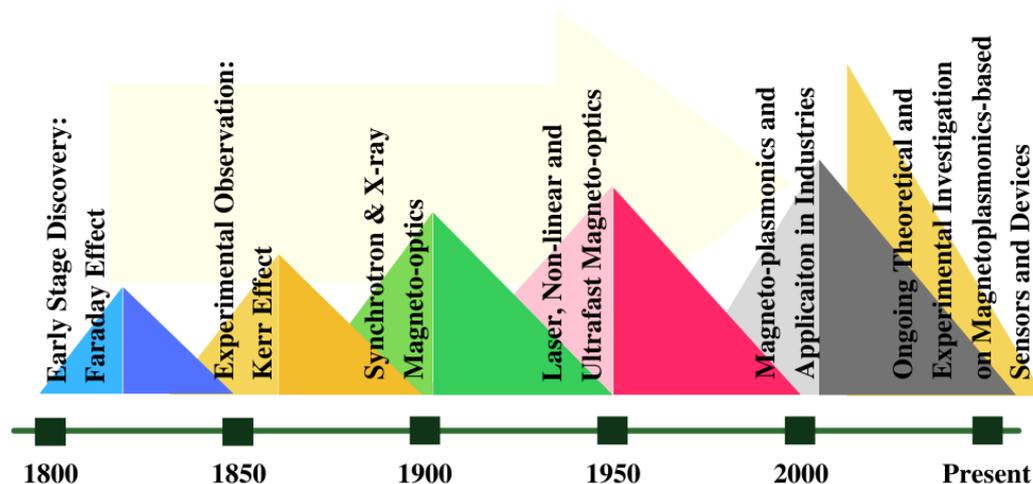

Figure 9. The origin of magneto-optics and emerging trends. 1800–1850: Early-Stage Discovery [1,145,146]: Faraday Effect; 1850–1900: Experimental Observation of the Kerr Effect [147]; 1900–1950: Synchrotron & X-ray Magneto-Optics [148]; 1950–2000: Laser, Non-linear and Ultra-fast Magneto-Optics [149]; and 2000–Present: Magnetoplasmonics [15,53,74,129,150,151], all-dielectric magnetophotonics [24,25, 32,33,35,42,124], and industrial Applications [70].

5. Concluding Remarks and Outlook

New MO materials and spectroscopy methods, with significant progress for on-chip integrability with reduced optical losses have been introduced during the last several years. In particular, amplified field amplitudes in plasmonic and all-dielectric magnetophotonic nanostructures have been exploited to improve sensing performance (higher sensitivity and resolution, with enhanced limits of detection), MO modulation of optical signals (in integrated optoelectronic circuits), and magnetic control of nanoparticle trapping, among other things. MO effects are now being used for the active modulation of optical chirality in structures ranging from nanoparticle arranges to chiral metasurfaces, which may open avenues for new, efficient, cheap, and all-optical enantioseparation of drugs in the pharmaceutical industry [70]. A range of potential applications that we identified are illustrated in Figure 10. Significantly, coupling together magnetic and plasmonic properties could

provide new routes for the separation of specific biological moieties, drug delivery, and photo/magnetothermal therapies. In fact, magnetoplasmonic nanotools that act as biosensors as well as theranostic agents have already been demonstrated [152], merging SERS (surface enhanced Raman scattering) with magnetic separation, but further investigations are necessary to integrate magnetoplasmonics with microfluidics technology [153] to bring these achievements from specialized laboratories to real-life applications.
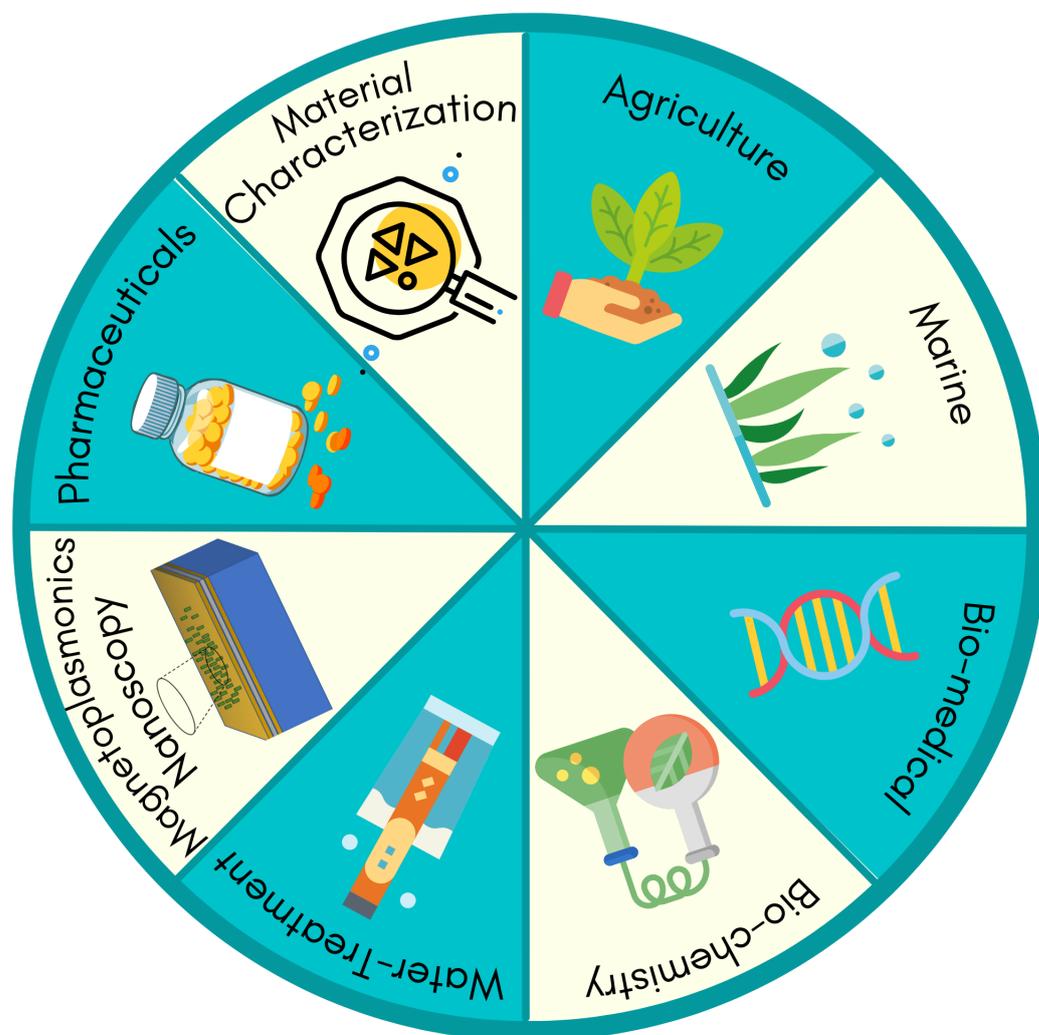

Figure 10. Potential applications of MO-SPR sensors in industry (e.g., nanostructure characterization, and label-free chemical and biological sensing) including but not limited to the following sectors: agriculture [154,155], marine [156], medical science [32,157], bio-chemistry [158], meteorology [159], nanoscopy [7–10,160], and pharmacy [70,106,161,162].

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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