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ABSTRACT

Plasmonics represents a unique approach to confine and enhance electromagnetic radiation well below the diffraction limit, bringing a huge potential for novel applications, for instance, in energy harvesting, optoelectronics, and nanoscale biochemistry. To achieve novel functionalities, the combination of plasmonic properties with other material functions has become increasingly attractive. In this Perspective, we review the current state of the art, challenges, and future opportunities within the field of magnetoplasmonics in confined geometries, an emerging area aiming to merge magnetism and plasmonics to either control localized plasmons, confined electromagnetic-induced collective electronic excitations, using magnetic properties, or vice versa. We begin by highlighting the cornerstones of the history and principles of this research field. We then provide our vision of its future development by showcasing raising research directions in hybrid magnetoplasmonic systems to overcome radiation losses and novel materials for magnetoplasmonics, such as transparent conductive oxides and hyperbolic metamaterials. Finally, we provide an overview of recent developments in plasmon-driven magnetization dynamics, nanoscale opto-magnetism, and acousto-magnetoplasmonics. We conclude by giving our personal vision of the future of this thriving research field.

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INTRODUCTION

The ability to control materials at the nanoscale allows to confine light into subwavelength volumes by exploiting collective electromagnetic-induced electronic excitations, known as plasmon polaritons.^{1–6} Unlike conventional optics, plasmonics enables unrivaled confinement and enhancement of the electromagnetic (EM) radiation well below the diffraction limit, containing a huge potential for real-life applications in energy harvesting, wave-guiding and lasing, optoelectronics, and nanoscale biochemistry.^{7–9} To achieve novel functionalities, the combination of plasmonic properties with other material functions has become increasingly appealing. In the past two decades, magnetoplasmonics in confined geometries has been an emerging area aiming to merge magnetism and plasmonics to control in new ways either the properties of localized plasmons using magnetic properties of matter or vice versa.^{10,11} In this Perspective, we review the current state of the art, challenges, and future opportunities in

some key areas of magnetoplasmonics. We begin with the use of localized surface plasmons (LSPs) to enhance magneto-optical (MO) effects in metallic systems, in the framework of active flat-optics metamaterials for light polarization control, lasing, as well as biochemical sensing, highlighting its history cornerstones and main principles and providing our vision of its future development.

We then venture into the new trends in the field. Relevant examples are the exploration of lossless and/or more exotic materials, such as plasmonic transparent conductive oxides (TCOs) and hyperbolic metamaterials (HMMs). We also briefly survey the latest advances in plasmon-driven magnetization dynamics and optomagnetism, e.g., the enhancement of the inverse Faraday effect in nanoscale geometries and the use of orbital angular momentum of light to achieve a superior ultrafast control of magnetism and spintronics.

We conclude by giving our outlook on the future of the field, showcasing new possible directions to achieve a full control of MO

effects and their enhancement by using nanoscale materials as well as to drive magnetic phenomena with plasmons at the atomic and sub-femtosecond timescales.

MAGNETOPLASMONICS ENABLED BY LOCALIZED PLASMONS

The recent development of nanophotonics has allowed for active and flexible light control and enabled numerous technological applications, such as metasurface polarization switches,^{12,13} chirality-sensitive nanoantennas,¹⁴ high resolution imaging and high sensitivity molecular detectors,^{15–17} and single-photon integrated quantum circuits.¹⁸ MO effects can offer the active and nonreciprocal control of the polarization and intensity of reflected and transmitted light. Therefore, the integration of MO-active materials in nanophotonics is particularly appealing for a variety of applications: ultrathin devices for optical isolation and modulation,^{19–21} information processing and cryptography,²² sensing,^{23–25} and metrology,^{26–28} including the ultra-sensitive detection of fluctuations in ferromagnetic meta-atoms caused by deep-subwavelength disorder via the photonic spin Hall effect,²⁹ spectroscopy,³⁰ imaging,³¹ structured light and holography,^{32–34} and spintronics.^{35,36}

Unfortunately, MO materials do not display sufficiently large MO effects when downsized to a sub-micron scale (e.g., Faraday rotation of a 1- μm thick Ce:YIG is only 0.5° in the infrared³⁷) and this has hindered the utilization of nanoscale magneto-optics in, for instance, active nanophotonics or light-driven nanomagnetism and spintronics, where a major goal is a coherent control of nm-sized magnetic bits.^{38,39}

A solution to address this challenge is represented by the combination of magnetic and plasmonic properties at the nanoscale. However, magnetoplasmonics can also be implemented exploiting propagating surface plasmon polaritons;^{11,40–44} hereafter, we will exclusively refer to the use of the peculiarities of optics at the nanoscale arising from the excitation of localized surface plasmons (LSPs) in metallic magnetic nanomaterials to enhance MO effects. Several-fold enhancement of magnetoplasmonic effects was indeed reported in nanostructures composed of pure ferromagnetic (FM) nanoantennas,^{21,41–43,45–48} while noble metal nanostructures display two orders of magnitude weaker magnetoplasmonic response at magnetic fields of the order of the Tesla.^{49–52} Even so, the LSP-mediated enhancement of MO is not enough to enable practical applications. The only area in which currently magnetoplasmonics can be advantageously applied is molecular and refractive index sensing.^{24,25,45,50,53–55}

According to a semi-classical theoretical description,⁴⁷ the enhancement of the MO activity in a magnetoplasmonic nanoantenna resonator, compared to a film of the same material and the same thickness, is understood as due to a second spin-orbit coupling (SOC) induced LSP (magneto-optical LSP, MO-LSP) driven by the LSP directly excited by the incident linearly polarized light. These two orthogonal LSPs display a phase lag and different amplitudes (see Fig. 1). The far-field MO response of the magnetoplasmonic nanoantenna results then from the ratio between the electric dipoles associated with the induced SOC-LSP and the directly excited LSP. In the case of a cylindrical magnetoplasmonic nanoantenna, the amplitudes of the LSP and MO-LSP electric dipoles are proportional to Q and Q^2 , respectively, where Q is the quality factor of the resonance. Ultimately, the MO activity of a magnetoplasmonic nanoantenna is Q -times larger

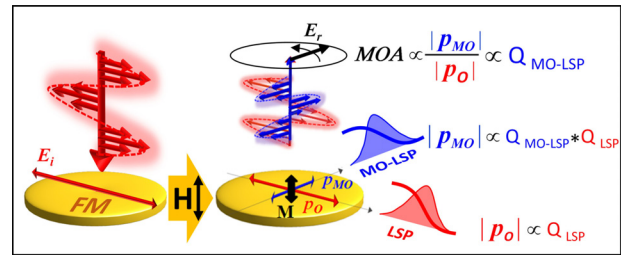


FIG. 1. Semi-classical model describing the interaction of linearly polarized light with a magnetoplasmonic nanoantenna. The figure sketches the electro-dynamics of a ferromagnetic (FM) disk generating an electric dipole (p_O) resonantly triggered by the electric field E_i of an incident linearly polarized electromagnetic radiation and a magneto-optically activated (via spin-orbit coupling) electric dipole (p_{MO}) by the application of a magnetic field H inducing a magnetization M in the nanoantenna. The enhancement of the amplitude of p_O at resonance is proportional to the Q-factor of the resonance along the direction of E_i (QLSP), while that of p_{MO} , induced by p_O and orthogonal to it, is proportional to the product of the QLSP and the Q-factor of the resonance in the orthogonal direction (QMO-LSP). The enhancement of the amplitude of the polarization change induced in the re-emitted electromagnetic radiation (magneto-optical activity) is proportional to the ratio between the module of p_{MO} and p_O and, therefore, limited to a factor QMO-LSP despite the much larger enhancement of p_{MO} . Readapted with permission from Fig. 1 of Ref. 47.

than that without LSP (e.g., that of a continuous film of the same thickness and material of the nanoantenna).

The quality factor Q is governed by the absorption and radiation losses, and different strategies have been explored to reduce such losses and achieve high values of Q . Absorption losses are particularly high in FMs, which result in a very small Q for pure FM nanoantennas (on the order of 2–5 to be compared to values on the order of 10 for noble metals in the same spectral range). While the search of magnetic materials other than metals (pure FM, non-magnetic (NM) materials and alloys) will be discussed in New Directions From a Materials Point of View section, here we focus on hybrid structures made of FM and noble metals. Much effort has been devoted in devising such hybrid structures, which chiefly contribute to the reduction of absorption losses bringing the Q -factor closer to that of a pure noble metal nanoantenna.^{19,56–61} Interestingly, such hybrid systems have been recently reported to enable lasing effects, which can also be tuned by using external magnetic fields.^{62,63}

The suppression of radiation losses in specific frequency ranges can be achieved by arranging plasmonic nanoantennas in a periodic array, thanks to the diffracted coupling of the EM field radiated by each nanoantenna. Diffractively coupled LSP is referred to as surface lattice resonance (SLR),^{64–67} and its excitation contributes to the increase in the Q -factor of the collective resonance of the array. By employing plasmonic SLRs in magnetoplasmonic arrays (viz., magnetoplasmonic crystals), a rich variety of interference features and further enhancement of MO activity have been reported in the last decade.^{59,68–71}

Despite all the effort devoted to increase the Q -factor, the achieved amplification of the MO effect is still not enough to enable applications. It is worth noting that the SOC-LSP electric dipole is proportional to Q^2 , which, upon reduction of absorption and radiation losses applying the strategies described above, can be of the order of 100 or even higher.⁷² The fact that the MO response is proportional to the ratio of the MO-LSP and LSP electric dipoles limits its

enhancement only to a factor Q , and thus, it is limited to 10–20 even upon reduction of losses. Physically, this can be understood as due to the simultaneous amplification of the re-emitted light by the LSP with the incident polarization. If the enhancement of the re-emission of light with the original polarization could be contained, amplifications of the MO effect up to Q^2 could be achieved. A recent study has shed light on this hidden amplification potential.⁷² It relied on non-concentric nanocavities consisting of a NM ring resonator near-field coupled to a FM disk. The near-field coupling induces the hybridization of the electric dipole of the FM disk with a multi-polar dark mode of the surrounding NM ring, leading to a Fano-like resonance for the hybrid mode.^{73,74} The concept is illustrated in Fig. 2. The result of the coupling leads to two key effects: (i) the electric dipole induced in

the FM disk is even larger than the LSP of the isolated disk; (ii) the multipolar mode of the NM ring acquires an electric dipole component that is oscillating out of phase with that induced by the incident light in the FM disk. The combination of these two effects benefits the MO activity enhancement in two ways: (i) effect corresponds to an increase in the LSP dipole amplitude, and thus of the SOC-LSP, beyond the Q value of the FM disk alone, and (ii) effect allows for the management, in particular, the reduction, of the radiated light with the original polarization of the incident field, thereby enabling a MO enhancement larger than Q that can eventually approach the theoretical Q^2 limit. For instance, the magnetoplasmonic nanocavity studied in Ref. 72 exhibited sevenfold enhancement in MO activity compared with that produced by the excitation of a LSP in the bare FM disks.

NEW DIRECTIONS FROM A MATERIAL POINT OF VIEW

Enabling a MO enhancement larger than Q seems very promising, and further research should consider the following criteria: low dissipative materials (NM/magnetic hybrids,^{20,60,72,75} dielectrics,^{76,77} and conductive oxides^{78,79}), small cavity footprints (higher surface coverage and MO effects), functional arraying (SLRs⁶⁷ and metagratings⁸⁰), anisotropic superlattice films (Au/Co, Au/Fe, Pt/Co⁸¹), epsilon-near-zero (ENZ) and near-zero-index (NZI) materials,^{82–84} and hyperbolic metamaterials.^{85–88} Much effort in the design of NM/FM hybrids has been reported by designing hybrid nanostructures made of noble metals (Au or Ag) and iron oxides or magnetic metals. Coating Au nanospheres (8 nm) with a thin FeO_x shell (1 nm) was found to enhance the MO response up to 50% with respect to uncoated Au nanospheres, which was ascribed to a purely dielectric effect.⁸⁹ However, if the volume of the iron oxide shell is increased, the MO response is fully dominated by the MO signal of the iron oxide moiety, masking any magnetoplasmonic effect.^{90,91} On the contrary, improvement of the magnetoplasmonic response was observed in different combination of materials, such as Ag@FeCo⁹² and FeO_x@Au⁹³ NPs prepared by chemical methods or sandwich-like Au/Co/Au nanodisks systems prepared by lithographic techniques.^{94,95} However, the enhancement of the MO response is not sufficient to exceed the one of pure ferromagnetic nanodisks, which remains the state-of-the-art material for nanoscale magnetoplasmonics. While strategies based on NM/FM hybrids and dielectrics have been widely reported and studied in the recent decades,^{11,96} here we would like to highlight two emergent classes of materials, which can contribute to fuel the development of future exciting new directions in this thriving research field: plasmonic transparent conductive oxides and hyperbolic metamaterials.

As mentioned in Magnetoplasmonics Enabled By Localized Plasmons section, critical limitations for the enhancement of the magnetoplasmonic response from the material point of view depend on the Q -factor of the LSP. However, these arguments are valid only for standard metals, either purely noble like Au and Ag or FM. In recent years, several new plasmonic or photonic nanostructures have emerged, enriching the field of plasmonics and photonics: dielectric^{76,77,97} and plasmonic degenerately doped semiconductor nanoantennas^{98–102} are among the most promising systems, but have been scarcely explored for magnetoplasmonics, thus holding an unexploited potential. Among plasmonic degenerately doped semiconductors, TCOs represent a valuable material choice for magnetoplasmonics. In NM plasmonic nanoparticles, the magnitude of the magnetic

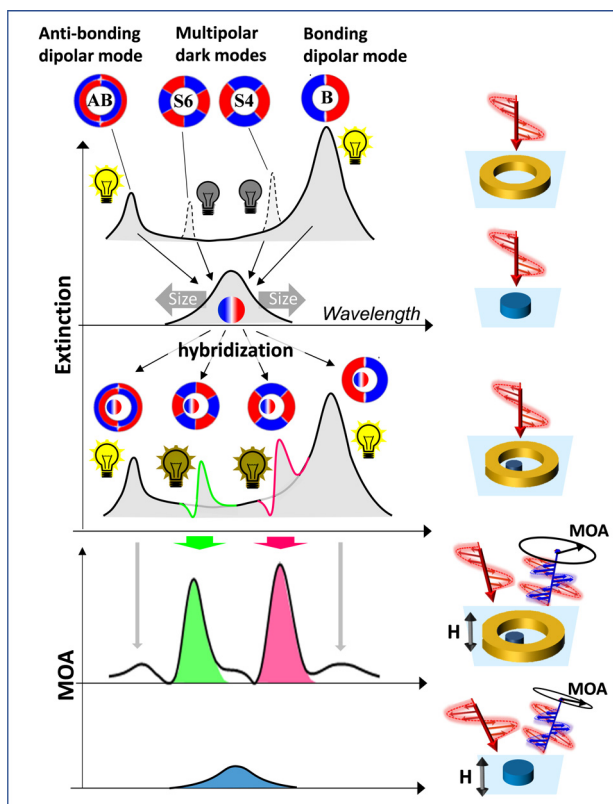


FIG. 2. Beyond the typical concept of LSP to enhance the MO response in confined geometries. General concept of hybridization with dark modes to boost the magneto-optical response illustrated utilizing the hybrid ring-dot structure reported in Ref. 72. The hybridization between the bright dipolar mode of the ferromagnetic dot with multipolar dark modes of a surrounding noble-metal ring results in Fano-like low radiant modes. The case of the hybridization with the hexapolar (S6) and quadrupolar (S4) modes is shown as an example. The excitation of one of these hybrid modes results in a strongly enhanced bright magneto-optical dipole in the ferromagnetic dot. The electromagnetic radiation with the incident polarization is re-emitted by the low-radiant hybrid Fano mode. The electromagnetic radiation with the polarization rotated by 90° is radiated by the enhanced bright magneto-optical dipole. Therefore, the magneto-optical activity, which results from the ratio of the two re-emitted EM fields, results strongly enhanced with respect to that achievable with a magnetoplasmonic antenna (e.g., a tenfold enhancement is reported in Ref. 72 compared to the results obtained with pure Ni disk nanoantennas in Ref. 47).

modulation is proportional to the cyclotron frequency ($\omega_c = qB/m$), which is inversely proportional to the effective mass of charge carriers (m). While in metals m is fixed to values close to the free electron mass m_e , in TCOs this value is typically much lower (0.2–0.3 m_e). Moreover, conductive oxides, such as ITO nanocrystals (NCs), display lower carrier densities than metals, shifting the plasmonic resonance in the near infrared, which is exploited for intriguing applications spanning from thermoplasmonics technologies^{103,104} to enhanced infrared spectroscopy.¹⁰⁵ Carrier charge can also vary if the carriers are either n- (electrons) or p-type (holes or oxygen vacancies): changing the sign of the charge in the carriers induces an opposite sign of the MO ellipticity.¹⁰⁶ Interestingly, in colloidal TCO-based NCs, the damping parameters can be considerably reduced through dopant choice and defect engineering, allowing to reach Q-factors values up to 12 in simple 10–20 nm spherical nanoparticles supporting electric dipole plasmonic modes.^{107–109} Radiative damping due to scattering is strongly reduced in TCO nanostructures having LSP above 1700 nm, as the scattering cross section for spheres is proportional to λ^{-6} and is, thus, strongly attenuated at such long wavelength (by approximately three orders of magnitude compared to nanostructures of the same volume having the LSP at 550 nm).¹¹⁰ A recent work demonstrated that colloidal TCO NCs enable to reach strong magnetoplasmonic response, challenging FM metallic nanostructures, due to the large Q-factor combined with the large cyclotron frequency (see Fig. 3).⁷⁹ The importance of keeping the plasmonic resonance sharp is highlighted by the stronger magnetoplasmonic response in FICO (F⁻ and In³⁺ doped CdO) with respect to ITO (Sn-doped In₂O₃). Proof of concept refractometric sensing experiments demonstrated the higher performances of TCO NCs also compared to ferromagnetic nanostructures (e.g., Ni nanodisks). However, in non-magnetic TCOs, such performance needed higher applied magnetic fields (around 1.4 T) with respect to Ni (0.2–0.3 T). While such applied field can be easily

achieved with hard disk writing heads,¹¹¹ the possibility to work at lower applied field would be preferable for magnetoplasmonic sensing devices in terms of costs and size of the final device.

To this purpose, the MO response of plasmonic TCOs could be further improved by introducing magnetic cations as co-dopants. Indeed, the emergence of free carrier-mediated magnetic interaction between magnetic cations was reported in thin films of magnetically doped TCOs, in some cases also reaching ferromagnetism at room temperature.¹¹² A clear example is represented by ZnO films doped with Co²⁺ or Mn²⁺ in the presence of free carriers.^{113,114} Such magnetic interaction can induce spin polarization of charge carriers, thus improving the magnetic modulation of the plasmonic response. Using nanostructures of these materials and tailoring their geometric and electronic features to achieve high Q-factors could potentially increase the coupling of free charge carriers with external magnetic fields, finally improving the magnetoplasmonic response of TCO nanostructures.

Recently, interesting correlations between localized plasmons and the MO response at the excitonic resonance have also been reported in doped semiconductor nanocrystals by Radovanovic *et al.*, where non-resonant coupling between excitons and the circular magnetoplasmonic modes has been observed, leading to a temperature independent MO signal in the bandgap region. Such effect results in the observation of the Zeeman splitting of the excitonic states in degenerately doped In₂O₃ NCs, which is not observed at room temperature in undoped NCs (where no carrier density is present in the conduction band), leading to a robust electron polarization at room temperature, opening interesting implications for spintronics and quantum information technologies.^{115–117}

Remarkably the large degree of tunability of charge carrier parameters (mass, charge, and density of the carriers) in these materials offers a great opportunity for boosting magnetoplasmonic devices performance close to those needed for active nanophotonic devices.

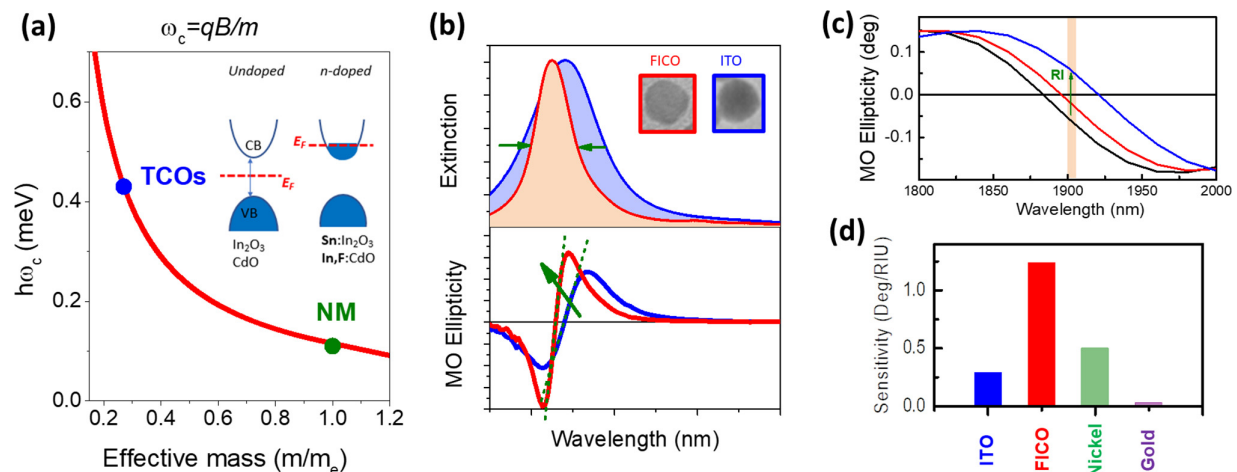


FIG. 3. Magnetoplasmonics beyond metals: transparent conductive oxides. (a) The cyclotron frequency (ω_c) is plotted as a function of the effective mass of charge carriers to show the main advantage of TCOs over noble metals (NM) for magnetoplasmonics. In the inset, the typical effect of aliovalent doping on the band structure of the TCO semiconductor is shown. (b) Extinction spectra and Faraday ellipticity of Sn-doped In₂O₃ (ITO) and F- and In-doped CdO (FICO) NCs, showing the effect of the reduced plasmon linewidth of FICO on the MO response, increasing the slope of the MO ellipticity close to the plasmonic resonance. (c) MO ellipticity measured at 1.4 T in three solvents with different refractive index (CCl₄, C₂Cl₄, and CS₂). (d) The MO refractive index sensitivity of ITO and FICO NCs, defined as MO ellipticity variation per refractive index unit at fixed wavelength is compared with nickel nanodisks and gold nanoparticles. Readapted with permission from Gabbani *et al.*, Nano Lett. **22**, 9036 (2022). Copyright 2022 Authors, licensed under a Creative Commons Attribution License.

For instance, in the context of sensing applications, it was already demonstrated that a combination of high-Q magnetic oxides and standard plasmonic materials can be very helpful for the detection of single analytes.¹¹⁸ Thus, the potential hold by TCOs can be exploited in a similar manner as well.

It is worth mentioning that ENZ and NZI materials, which display a phase refractive index close to zero at specific wavelengths, are known to enhance or inhibit light-matter interactions, thus representing a new frontier in magnetoplasmonics and magneto-optics. In fact, they can, for instance, allow for the excitation of additional plasmonic and/or photonic modes (such as strong and sharp bright magnetic dipolar modes) in confined geometries, which can be combined with the previously reported strategies to enhance Q beyond the current limits. Thus, they represent an additional and very interesting playground to explore new ways for enhancing MO activity at the nanoscale. A concrete example of this potential is represented by a sub-class of these materials, i.e., HMMs. One of the most interesting properties of these materials is that they display a strong optical anisotropy and nontrivial electro-optical properties, such as conductive behavior along particular spatial directions and insulating behavior along the others, even in nanoscale geometries like antennas or plasmonic crystals.^{85,86,119–123} In 2021, it was reported that NM HMM nanoantennas made of Au/TiO₂ multilayers can display an enhanced MO response driven by the hyperbolic dispersion via the coupling of either electric or magnetic dipolar optical modes with static magnetic fields [see Figs. 4(a) and 4(b)]. In particular, the magnetic dipole exhibits a much sharper resonance, thus, a much higher Q-factor. If we couple this mode with the multipolar mode of a nanoring, the results obtained in Ref. 72 would be further amplified, thus potentially approaching, and even exceeding, the theoretical Q² limit and perhaps unlock unforeseen MO effects. Finally, a remarkable example of application of these materials has been recently reported by Díaz-Valencia *et al.*,¹²⁴ who predicted that MO HMMs can be potentially much more sensitive to refractive index variation compared to common HMMs-based sensors, which can be still considered unbeaten [see Figs. 4(c)–4(e)].¹²⁵ This performance can be even improved if proper design rules to increase the quality factor of the modes are implemented.¹²³ Overall, even if the field of magnetoplasmonics is quite well established, based on these last developments from a materials viewpoint, we feel that we are just scratching its surface.

ULTRAFAST OPTO-MAGNETISM WITH PLASMONICS

An interesting emerging aspect in the field of magnetoplasmonics is the possibility to exploit MO phenomena on the nanoscale by using femtosecond (fs) light pulses. For instance, time-resolved MO spectroscopy makes it possible to probe spin dynamics at the timescale of fundamental magnetic interactions, that is the exchange and the SOC interactions, responsible for the magnetic order at the macroscale. Furthermore, the reciprocal interaction between spins and fs light pulses is one of the main topics in fundamental magnetism.¹²⁶ In particular, the subwavelength regime of light-spin interaction revealed the plasmon-mediated enhancement and emergence of new MO phenomena at ultrafast timescales.^{127,128} Novel possibilities of optical control of magnetism on the nanometer scale via plasmonic resonances have emerged as well.

After the discovery of fs demagnetization by ultrashort laser pulses in Ni films,¹²⁹ the ability to manipulate optical pulses at a timescale where we can achieve coherent interaction between photons,

charges, and spins, has opened the door to exploit the electric-field oscillations of light to control magnetism in unprecedented and unexpected ways.^{130–133} Ground-breaking experiments have shown that we can exploit the helicity (right or left circular polarization state) of light to act coherently on the spin moment of electrons,^{134–137} as well as induce transient MO effects in nonmagnetic media, where the instantaneous magnetization of the medium results from the light helicity-induced angular momentum of electrons.^{138,139} The observation of these effects fueled intense debates over which physical mechanisms drive the processes underlying the electron spin/charge-light helicity coupling, and several theories have been developed, from classical plasma models,^{140,141} up to quantum,^{142,143} *ab initio*,¹⁴⁴ and relativistic¹⁴⁵ descriptions.

All these groundbreaking experimental and theoretical studies on ultrafast opto-magnetic effects led to the development of the all-optical switching (AOS) of magnetization.¹⁴⁶ The first AOS of magnetization was demonstrated in a ferrimagnetic amorphous GdFeCo by a single 40 fs circularly polarized laser pulse. After careful examination, it turned out that the magnetic circular dichroism (MCD) and the different demagnetization processes between Gd and Fe sublattices enable this helicity dependent ultrafast (on the order of 30 ps) magnetization control.^{147–150}

Further studies have been reported, mainly using ferrimagnetic and ferromagnetic metallic systems, such as GdFeCo, TbFeCo, Co/Pt, and Fe/Pt.^{136,137,151,152} Although there are some differences in the opto-magnetization processes, every switching mechanism in the metallic systems can be attributed to laser-induced heating near the Curie temperature.^{150,153} A non-thermal AOS (i.e., AOS not dependent on laser-induced material heating) has been only demonstrated in a dielectric of Co-doped yttrium iron garnet (YIG:Co) through photo-induced magnetic anisotropy and showed ultrafast write-read time (<20 ps) together with unprecedentedly low heat load (<6 J/cm³).¹⁵⁴ The Co dopants exhibit strong single ion anisotropy depending on the valence of the ion, which can be modified by light-induced electronic transitions at the Co ions in nonequivalent crystal sites, and, thus, the magnetic anisotropy of the YIG:Co is controllable with a linearly polarized ultrashort pulsed laser source. Therefore, its AOS can be determined by the orientation of the linearly polarized light due to the crystalline symmetry.¹⁵⁵ The smallest domain size achieved in film samples of the studies mentioned above was approximately 5 μm. To meet the storage density over 1 Tbit/inch², the AOS will have to be achieved beyond the diffraction limit with the lowest possible energy. If the switching is achieved within a 20 × 20 × 10 nm³ domain, the AOS only needs 10 fJ in GdFeCo¹⁵⁶ [see Fig. 4(a)] and 3 aJ in YIG:Co,¹⁵⁴ approaching the minimum energy required for a stable bit at room temperature, 60 k_BT (0.25 aJ). This challenge has been attempted by either miniaturization of opto-magnetic materials by lithography^{137,151,156,157} or using plasmonic antennas.^{11,127,152} A nanostructured GdFeCo demonstrated a 200 nm domain AOS with a single fs light pulse.^{156,157} A few hundred nm long Au wires shaping similar to a dipole antenna on TbFeCo film successfully concentrate a pulsed laser with a wavelength of 1030 nm, and the AOS was achieved within ~50 nm in diameter [see Fig. 5(a)].¹⁵² In 2022, magnetoplasmonic crystal arrays composed of Au truncated cones topped with TbCo nanodisks on a glass substrate have exhibited plasmon-assisted thermal demagnetization.⁷¹ The authors not only demonstrated a threefold enhancement in the demagnetization efficiency at the localized plasmon resonance

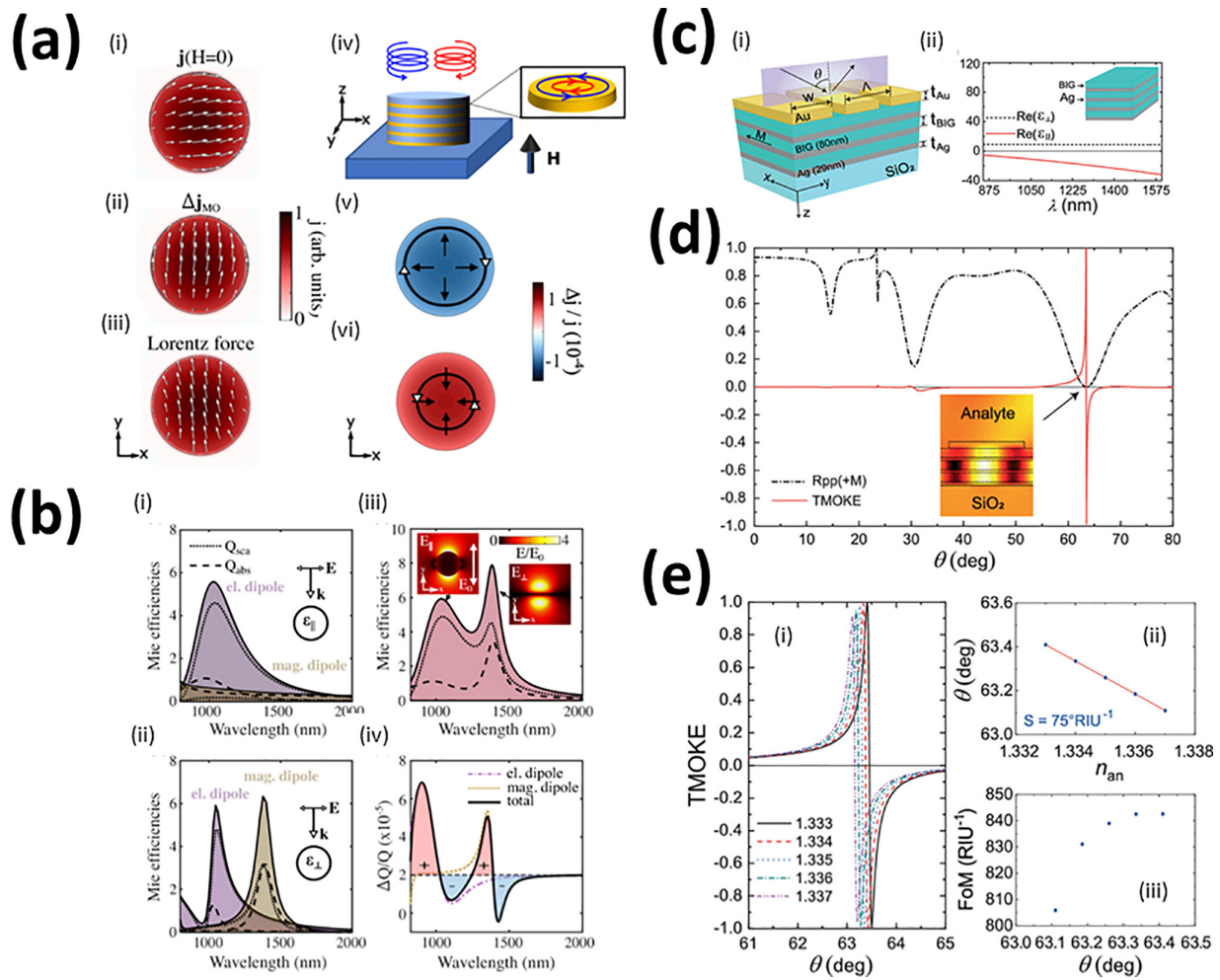


FIG. 4. Magnetoplasmonics beyond metals: hyperbolic metamaterials. (a) (i)–(iii) Calculated current density \mathbf{j} (white arrows) at the low-energy resonance for zero magnetic field (i), magnetically induced change of current $\Delta\mathbf{j}_{MO}$ (ii), and Lorentz force calculated as cross product of \mathbf{j} and the unit vector in direction of \mathbf{H} (iii). The absolute value of \mathbf{j} is color coded for all three cases. (iv) Circular dichroism effect in terms of spatial confinement or broadening of circular modes. (v) and (vi) Induced relative change of current density $\Delta\mathbf{j}/\mathbf{j}$ (color coded) for opposite helicity of the incoming electric field, as sketched in the plot. Black arrows show the gradient of $\Delta\mathbf{j}/\mathbf{j}$ and display the induced spatial broadening or confinement of the circular plasmonic motion. (b) Scattering (dotted lines) and absorption (dashed lines) efficiencies Q_{sca} and Q_{abs} for spherical nanoparticles with isotropic permittivity $\epsilon_{||}$ (i) and ϵ_{\perp} (ii). Interaction efficiencies are analytically calculated with the Mie expansion up to the first order, where magnetic (green) and electric (purple) contributions are separately evaluated. (iii) Sum of the in-plane electric dipole shown in (i) and the out-of-plane magnetic dipole shown in (ii). Inset are near-field plots of the resonances normalized to the incoming field E_0 indicated by the white arrow. (iv) Differential extinction spectrum $\Delta Q/Q$ calculated from the magnetically induced dichroism effect. Contributions from the electric and magnetic dipoles are marked by the dash-dotted and dashed lines, respectively. The overall spectrum (black solid line) exhibits a derivative spectral shape at the resonances, as indicated by the light blue and light red areas under the curve. Readapted with permission from Kuttruff *et al.*, Phys. Rev. Lett. **127**, 217402 (2021). Copyright 2021 Authors, licensed under a Creative Commons Attribution License. (c) (i) Schematic representation of the proposed architecture, comprising alternating metal/dielectric (Ag/BIG) layers on a SiO_2 substrate. A periodic one-dimensional Au grating is used to couple the incident light to the hyperbolic bulk modes in the multilayer structure. (ii) Real part of the effective permittivity $\epsilon_{||}$ and ϵ_{\perp} , calculated for an infinite multilayer structure. (d) $R_{pp}(+M)$ and transverse MO Kerr effect (TMOKE, i.e., magnetic modulation of light intensity) for $\Lambda = 830$ nm vs incidence angle. The inset shows the normalized magnetic field component of the hyperbolic bulk mode. (e) TMOKE vs incident angle at various n_{an} . (b) Angle corresponding to the minimum peak of TMOKE as a function of n_{an} represented by solid circles. The solid line represents the linear fitting, whose slope gives $S = 75^\circ \text{RIU}^{-1}$. (c) FoM as a function of the angle associated with the minimum peak of TMOKE. Readapted with permission from Díaz-Valencia *et al.*, ACS Appl. Nano Mater. **5**, 1740 (2022). Copyright 2022 American Chemical Society.

but also showed its mitigation by using the SLR of the array [Fig. 5(b)]. A similar very promising approach was also reported by Vergès *et al.*, who demonstrated that both the single-pulse optical writing and magneto-optical readout are significantly enhanced by the excitation of collective SLRs in perpendicularly magnetized magnetoplasmonic

crystals.¹⁵⁸ Other strategies to achieve more efficient magnetization switching processes by exploiting plasmon-induced thermal effects were also recently reported.¹⁵⁹ Similar thermal effects can also strongly modulate MO effects (e.g., transverse MO Kerr effect),¹⁶⁰ as well as impact demagnetization processes on the ps and nm scales.¹⁶¹ The plasmonic

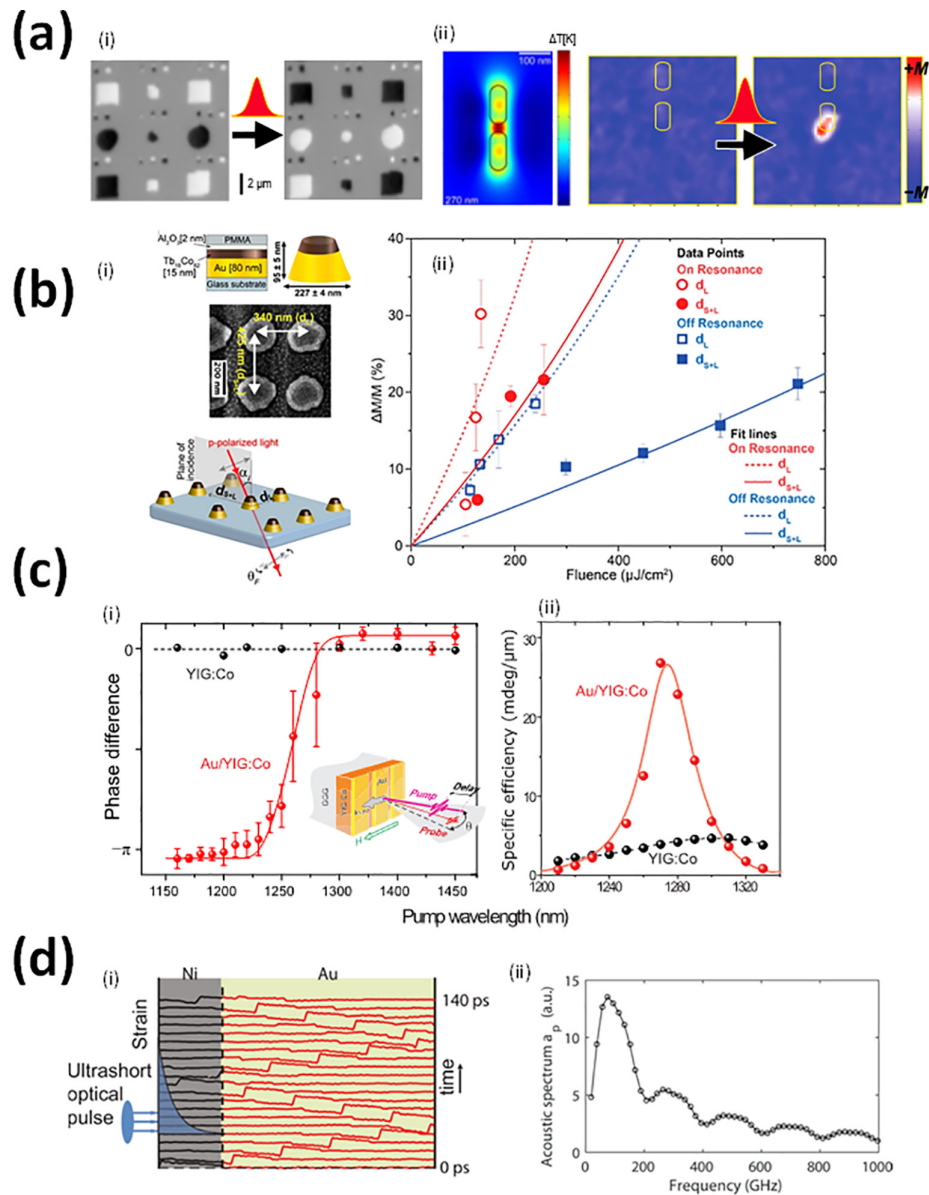


FIG. 5. Nanoscale control of magnetization dynamics with magnetoplasmonics. (a) (i) Magnetization switching by a single fs laser pulse demonstrated in out-of-plane domains with sizes down to 200 nm in GdFeCo nanostructures. Readapted with permission from Le Guyader *et al.*, *Appl. Phys. Lett.* **101**, 022410 (2012). Copyright 2022 AIP Publishing. (ii) Left: FDTD simulation of the temperature increases in the TbFeCo layer beneath the Au nanoantenna assuming a light pulse of 3.7 mJ cm^{-2} . Right: magnetic contrast images before and after a single laser pulse. Readapted with permission from Liu *et al.*, *Nano Lett.* **15**, 6862 (2015). Copyright 2015 American Chemical Society. (b) (i, top) Schematic of the nanoantenna structure and composition (i, middle) SEM of the magnetophotonic surface crystal with two periodicities along the perpendicular directions labeled d_{S+L} and d_L , and (i, bottom) experimental geometry for optical transmittance and magneto-optical spectra measurements indicating the plane of incidence and angle of incidence (α_i) relative to the surface normal. (ii) Peak values of demagnetization plotted as a function of applied fluence. The solid data points are for measurement along the d_{S+L} direction, and the open data points for measurement along the d_L direction. Solid lines are Curie law fits for data points along the d_{S+L} direction, and dashed lines are Curie law fits for data points along the d_L direction. Red data points/lines denote on-resonance pumping and blue data points/lines off resonance pumping of the localized plasmon mode. Readapted with permission from Mishra *et al.*, *Nano Lett.* **22**, 9773 (2022). Copyright 2022 Authors, licensed under a Creative Commons Attribution License. (c) In the Au/YIG:Co, the magnetic precession reversal (i) is seen across the plasmon resonance at $\sim 1270 \text{ nm}$ (i), where the photomagnetic efficiency is enhanced (ii) compared with the bare YIG:Co. Panel (i) is readapted with the permission from Kazlou *et al.*, *Appl. Phys. Lett.* **120**, 251101 (2022). Copyright 2022 AIP Publishing. Panel (ii) is readapted with the permission from Kazlou *et al.*, *ACS Photonics* **8**, 2197 (2021). Copyright 2021 Authors, licensed under a Creative Commons Attribution License. (d) (i) Ultrafast optical excitation of a free-standing Ni/Au bilayer generates ultrashort, picosecond strain pulses $\varepsilon_{zz}(z,t)$ bouncing back and forth in the layer. (ii) The acoustic spectrum for a 30 nm Ni/70 nm Au bilayer obtained using the matrix inversion protocol. Reproduced with permission from Vernik *et al.*, *Phys. Rev. B* **106**, 144420 (2022). Copyright 2022 American Physical Society.

enhancement of photo-induced spin precession was reported for YIG:Co with a 1D Au grating. From the comparison with a bare YIG:Co, the amplification efficiency was estimated to be sixfold at the resonance with the field confinement of 300 nm in the film [Fig. 5(c)].^{162,163} A similar enhancement was also observed in the inverse Faraday effect in a Gd-Yb-doped bismuth iron garnet (GdYbBIG) covered with a 1D Au grating, and the excitation efficiency of subterahertz spin precession was resonantly enhanced by two orders of magnitude at the resonance within 100 nm deep in the film.¹⁶⁴ The main function of the plasmon structures in the above applications is to concentrate a laser light. However, it is noteworthy that the same Au/YIG:Co structure as in Ref. 163 exhibits an abrupt phase reversal of the magnetization precession at the plasmon resonance.¹⁶⁵ Since the photo-induced magnetic anisotropy is described by orthogonal electric field components,¹⁵⁴ this is presumed to be the result of the phase shifts in the EM field at the plasmon resonance and the interference between the plasmonic field and the incident field. Although the azimuthal angle dependence of the phase shift in photo-induced spin precession has been reported for a bare GdYbBIG and explained by both the inverse Cotton-Mouton effect and photo-induced magnetic anisotropy,¹⁶⁶ the one caused by plasmonic resonance is not fully understood yet. Therefore, this finding has revealed another nontrivial role of plasmons in opto-magnetism and suggests new routes to spin manipulation by light with plasmonics.

All the above considerations are built upon exploiting only the helicity (right or left circular polarization) of light. An optical beam can carry also orbital angular momentum (OAM).^{167,168} Combining OAM with plasmonics, sub-fs dynamics of OAM can be realized in nanoplasmonic vortices,^{32,169–171} which can be confined to deep subwavelength spatial dimension and could offer an excellent time resolution. Thus, the OAM is expected to have an impact also on magnetoplasmonics, where it could offer a new approach to control magnetic phenomena.^{32,38} Recently, observations of an interaction between magnetism and beams carrying OAM were reported.^{34,172} By using a hydrodynamic model, Karakhanyan *et al.* numerically quantified the relative contributions of helicity and OAM of light to the inverse Faraday effect (IFE) in a thin gold film illuminated by different focused circularly polarized beams carrying OAM.¹⁷³

The OAM of light provides a new degree of freedom to control the IFE, and the resulting opto-magnetic field has the potential to influence numerous research fields, such as all-optical magnetization switching and spin-wave excitation. These findings support a mechanism of coherent transfer of angular momentum from the EM field to the electron gas and pave the way for new strategies for optical isolation that do not require externally applied magnetic fields. We foresee that exploiting the OAM of light in plasmonic structures will become paramount for the future development of magnetoplasmonics. As of now, OAM has been exclusively experimentally utilized for metrology, as a probe of magnetic textures and dynamics.^{34,172} The use of OAM for the optical manipulating magnetization, e.g., in ultrafast AOS experiments, remains an almost uncharted territory, which, based on preliminary works,^{175,176} holds great promises and opportunities for the future.

Overall, plasmonics can make ultrafast opto-magnetism at the nanoscale more fascinating for probing and controlling the dynamics of both charges and spins. The unprecedented speed and low dissipations of the AOS together with plasmonic localization can be utilized to accommodate ever increasing demand for data storage. It is worth

adding that for the miniaturization of thermally stable magnetic domains, not only the localization of the EM field but also the development of opto-magnetic materials with relatively high anisotropy is important. Although the thermal ultrafast opto-magnetic effects are quite well understood, much effort to shed more light onto the origin of fundamental opto-magnetic effects is currently devoted to broadening experimental and theoretical studies well into a timescale (10–100 fs, nonthermal regime) where nonthermalized electrons and plasmon dephasing effects can play a major role. Overall, the study of plasmon dephasing effects on the spin dynamics in MO materials is still an unexplored terrain. This opportunity can open new perspectives in both fundamental and applied aspects of ultrafast magneto-plasmonics. For instance, by working in the sub-100 fs timescale, i.e., when electrons are not yet thermalized, we can control spin dynamics also with metallic nanostructures, thus overcoming the intrinsic limitations placed by Ohmic losses and maybe reaching higher Q-factor on the fs timescale, opening excellent opportunities toward plasmon-driven MO and magnetic functionalities with a working bandwidth of hundreds of THz.³⁸

Finally, it is worth mentioning that, without any need to use plasmonic structures, it is already possible to use fs light pulses to generate magnons with nm wavelengths.^{127,177} An interesting perspective in this direction is offered by the exploitation of artificial freestanding metal-ferromagnet multilayer magneto-phononic cavities to generate a strong phonon-magnon coupling, to excite perpendicular standing spin waves exchange magnons up to 1 THz [see Fig. 5(d)].¹⁷⁴ Moreover, if we think about the emerging possibilities enabled by plasmonic picocavities,¹⁷⁸ where the EM field oscillations can be controlled on the pm-scale, we can foresee that in the near future, it might be possible to exploit these pico-cavities to excite “atomic-scale” plasmons¹⁷⁹ to control magnetism at the atomic scale with extreme (sub-nm) resolution. By leveraging also on single electron control with plasmonic antennas,^{180,181} it will be even possible to unlock the door for driving the spin of single electrons with sub-fs resolution. Ultrafast nonthermal magnetoplasmonics represent an intriguing pathway to control ultrafast spin dynamics at the fs and nm scales, and we foresee that this is just the beginning of a new chapter in the fascinating story of magnetoplasmonics.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Nicolò Maccaferri: Conceptualization (equal); Supervision (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Alessio Gabbani:** Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Francesco Pineider:** Conceptualization (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Terunori Kaihara:** Validation (equal); Writing – original draft (equal); Writing – review & editing (equal). **Tilaike Tapani:** Validation (equal); Writing – review & editing (equal). **Paolo Vavassori:** Conceptualization (equal); Project administration (equal); Supervision (equal); Validation (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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