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# Anisotropic Nanoantenna-Based Magnetoplasmonic Crystals for Highly Enhanced and Tunable Magneto-Optical Activity

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**(5)** Supporting Information

**ABSTRACT:** We present a novel concept of a magnetically tunable plasmonic crystal based on the excitation of Fano lattice surface modes in periodic arrays of magnetic and optically anisotropic nanoantennas. We show how coherent diffractive far-field coupling between elliptical nickel nanoantennas is governed by the two in-plane, orthogonal and spectrally detuned plasmonic responses of the individual building block, one directly induced by the incident radiation and the other induced by the application of an external magnetic field. The consequent excitation of magnetic field-induced Fano lattice surface modes leads to highly tunable and amplified magneto-optical effects as compared to a continuous film or metasurfaces made of disordered noninteracting magnetoplasmonic anisotropic nanoantennas. The concepts presented here can be exploited to design novel



magnetoplasmonic sensors based on coupled localized plasmonic resonances, and nanoscale metamaterials for precise control and magnetically driven tunability of light polarization states.

**KEYWORDS:** Magneto-optical Kerr effect, lattice surface modes, plasmonic anisotropy, magnetoplasmonics

etallic nanostructures are known to exhibit collective electronic oscillations, so-called localized surface plasmon resonances (LSPRs), which determine their optical response in the visible and near-infrared spectral range and lead to extreme confined and enhanced electromagnetic fields at the nanoscale.<sup>1-4</sup> Since the past decade, conventional metallic materials, such as gold and silver, have been used for creating electromagnetically coupled nanostructured systems, dubbed metamaterials, with novel optical properties emerging from the subwavelength confinement of light.<sup>5</sup> The optical response of these systems can be tailored by designing the response of the individual units as well as the electromagnetic coupling between them. Besides its fundamental importance, manipulation of light at the nanoscale is of great interest in applied research fields for the prospect of applications in reallife,<sup>6</sup> such as energy harvesting and photovoltaics,<sup>7,8</sup> wave-guiding and lasing,<sup>9,10</sup> optoelectronics<sup>11</sup> and biochemistry.<sup>12,13</sup> Among the exciting phenomena arising from light manipulation at the nanoscale, an emerging perspective relies on the possibility to create multifunctional metamaterials, where the optical properties can be actively tuned by exploiting the synergistic interplay between different physical properties of the constituents. The plasmonic response can be modified for instance through the excitation and tuning of lattice phonons (acoustoplasmonics) or spins of the electron gas in the material

(magnetoplasmonics).<sup>14–16</sup> Among them, magneto-optical (MO) metamaterials based on magnetic plasmonic nanoantennas offer promising routes to actively manipulate light properties at the nanoscale using magnetic fields. The interplay between electronic charge oscillations and their magnetic response controlled by an external stimulus such as a static or dynamic magnetic field, has been already shown to bring novel and unexpected optical effects.<sup>17,18</sup> In more detail, pure ferromagnetic nanoantennas support LSPRs and display a magnetic field-induced optical activity, namely a MO activity (MOA), which can be exploited to achieve an active and remote magnetic field control of light polarization over the entire visible and near-infrared spectral regions.<sup>19-21</sup> It has been recently shown that the field-induced MOA of pure ferromagnetic nanoantennas can produce localized plasma oscillations along directions perpendicular to the LSPR that is directly excited by the incident light, owing to the inherent spin-orbit coupling (SOC) in the constituent material. For instance, a radiation impinging normally on a ferromagnetic nanoantenna results in the excitation of two mutually orthogonal localized plasmon oscillations lying in the plane

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**Figure 1.** (a) Left panel: schematic of a MPC made up of 30 nm thick nickel nanodisks with pitch a = 500 nm. (a) Right panel:  $Q_{ext}$  spectra of the MPC sketched in the left panel with the diameter D of the nanodisks increasing from 100 nm to 200 nm (black curves). The red curves indicate the LSPR of an isolated nanodisk. (b) Left panel: schematic of the system studied in this work, where the single nickel nanoparticles displays two different optical responses along the two in-plane principal *x*- and *y*-axis, aligned with the nanodisk LA = 180 nm and SA = 100 nm, respectively. The thickness of the single antenna is 30 nm. (b) Right panel:  $Q_{ext}$  spectra of the MPC sketched in the left panel (black curves). The red curves indicate the LSPR of an isolated nanodisk. The vertical green solid lines indicate the spectral position of the Rayleigh's anomaly  $\lambda_R$ . The system is assumed to be immersed in a homogeneous refractive index n = 1.5. All the curves were calculated using the Coupled Dipole Approximation (CDA).

perpendicular to the radiation wavevector, when a magnetic field is applied parallel to the propagation direction of the incident radiation. Besides its potential application to flat-optics nanodevices, the MO-induced optical activity of magnetic antennas has been employed to design ultrasensitive refractive index and molecular detectors,<sup>22</sup> as well as magnetically controllable rulers with an extreme sensitivity to nanoscale distances.<sup>23</sup> On the other hand, pure ferromagnetic nanoantennas with appreciable MOA are made of transition metals (e.g., nickel and cobalt), which display larger dielectric losses compared to conventional metals used in plasmonics.<sup>24–26</sup> The intrinsic plasmonic damping of the constituent material results in broader and less intense LSPRs. To overcome the problem, various strategies have been applied so far, such as the fabrication of complex hybrid noble-metal/ferromagnetic structures or inducing coupling of broad localized modes with narrow excitations such as surface plasmon polaritons.<sup>27-33</sup> Likewise, plasmonic crystals (PCs) based on ordered arrays of metallic antennas were shown to produce an exceptional sharpening of plasmonic resonant features. The presence of a periodic lattice structure gives rise to a diffractive coupling through the far-fields scattered by the nanoantennas at particular wavelengths dictated by the geometry and periodicity of the PC. This leads to the excitation of collective lattice surface modes (LSMs) showing Fano line shapes.<sup>34-44</sup> MO

properties of periodic arrangements of MO-active antennas are starting to attract considerable attention. In particular, periodically arranged circular ferromagnetic nanoantennas have been studied very recently with the aim to exploit Fano LSMs to overcome the aforementioned limitation of the high losses and obtain narrow and intense MO resonant response.<sup>45,46</sup> While the effects of lattice geometries and nanoantennas size and shape on the excitation of LSMs in noble-metal based PCs has been studied extensively,<sup>34–44</sup> their effects on the MO properties of PCs made of pure ferromagnetic elements are still unexplored.

Here we generalize and broaden the implementation of PC concept to the magnetoplasmonics framework to design a magnetoplasmonic crystal (MPC) based on the excitation of anisotropic LSMs. We first show how the resonant excitation of in-plane LSMs can be conveniently and finely controlled by the anisotropic in-plane plasmonic response of the individual building blocks of the MPC, namely ferromagnetic elliptical nanodisks. Then, we demonstrate that the activation of a magnetic field induces a second LSPR in the single disk, spectrally separated and orthogonal to that directly excited by the incident light. This results in the simultaneous excitation of two perpendicular LSMs, one optically induced and the other MO-induced, leading to an enhanced and highly tunable MOA. In more detail, we demonstrate that the enhancement and



**Figure 2.** (a) SEM image of a portion of the elliptical nanoantennas on glass randomly distributed (top) and in the MPC (bottom). SA = 100 nm, LA = 250 nm, and thickness t = 30 nm. The pitch of the MPC is 400 nm in both LA and SA directions and the filling factor of both samples is 12%; scale bars 500 nm. (b) Left panel: sketch of the system studied when  $E_{inc}$  is applied along the LA (red incident radiation) and the SA (blue incident radiation). The solid arrows inside the particles represent the oscillation of the excited dipoles whereas the ones outside indicate the direction of the predominant far-field interaction in the explored spectral range. (b) Right panel: experimental  $Q_{ext}$  spectra measured for a system of randomly arranged elliptical antennas (dashed lines) and for the MPC (solid lines) for both SA-(blue lines) and LA-(red lines) illumination.  $\Delta Q$  for the random (violet dashed line) and MPC (violet solid line) case. (c) Left panel: sketch of the same system shown in (b) when an external static magnetic field is applied along the direction perpendicular to the lattice plane and for  $E_{inc}$  applied along the SA (blue incident radiation) and LA (red incident radiation). The SOC activated by the magnetic field induces dipolar oscillations perpendicular to the direction of the incident illumination (MO-LSPRs), leading to a predominant far-field interaction parallel to the polarization of  $E_{inc}$  (MO-LSMs). (c) Right panel: experimental MOA spectra measured for a system of randomly arranged elliptical antennas (dashed lines) and for the MPC antenas (dashed lines) and for the magnetic field interaction parallel to the polarization of  $E_{inc}$  (MO-LSMs). (c) Right panel: experimental MOA spectra measured for a system of randomly arranged elliptical antennas (dashed lines) and for the MPC (solid lines) for both SA-(blue lines) and LA-(red lines) illumination.  $\Delta_{MOA}$  for the random (violet dashed line) and MPC (violet solid line) cases.

tunability of the MOA of the MPC can be strongly amplified and precisely molded around selected wavelengths through the line shape tuning of the LSMs, achieved via the proper design of the shape and size of the individual antennas and of the periodicity of the lattice. This study sheds light on how to advantageously steer the synergistic combination of plasmonic anisotropy of the individual magnetic nanoantennas building the MPC and their diffractive coupling, leading to a far richer physics and exploitable phenomenology of magneto-optical effects in nanoscaled magnetoplasmonic materials.

We first analyze the key mechanism that governs the dependence of LSMs on the size of the individual building blocks of the MPC, while the period of the lattice, assumed to be square, is fixed. In the schematic shown in the left panel of Figure 1a the electric field  $E_{inc}$  of normally incident radiation is assumed to oscillate either along the principal *x*- or *y*-direction of a MPC made of plasmonic nanoantennas with in-plane aspect ratio (AR) equal to 1, that is, circular disk-shaped antennas. We consider that the nanoantennas have an edge-to-edge distance large enough (>100 nm) that we can neglect

near-field interactions in the visible and near-infrared spectral region (see Supporting Information for detailed discussion and Figure S1). At these frequencies, the electric field of the incident radiation induces dipolar LSPRs in each nanoantenna along its oscillation direction. LSMs arise from the onset of farfield in-plane diffractive coupling between the fields scattered by the individual antennas, according to the criterion developed by Rayleigh.<sup>47</sup> In the spectral region of interest (550-1100 nm), the strongest diffractive interaction occurs in the in-plane direction perpendicular to that of the incident electric field, at a wavelength  $\lambda_{\rm R} = a \cdot n$  ((1,0) diffraction modes), where *a* is the array pitch in such a perpendicular direction and n is the refractive index of the surrounding medium. The wavelength  $\lambda_{\rm R}$ corresponds to the photon energy at which the wavelength of the radiation scattered by the individual antenna is commensurate with the periodicity of the array and is related to the incipient appearance of the first order diffracted radiation in the free space.<sup>36,38</sup> Depending on the relative position between the LSPR of the individual disk and  $\lambda_{R}$ , the diffractive coupling between the radiation fields produces Fano LSMs with different line shapes.<sup>38</sup> This is clearly shown in the calculated extinction efficiency Qext spectra plotted in the right panel of Figure 1a, where we varied the diameter D of circular nickel disks, arranged in a periodic fashion (a = 500 nm along both xand v-direction), from 100 to 200 nm. The thickness of the disks is 30 nm. The plotted curves were calculated using the coupled dipole approximation (CDA) method (see Supporting Information for a detailed derivation), which is known to give an excellent description of the optical response of this kind of systems and it is based on the assumption that the individual antenna radiates as a point-dipole.<sup>36</sup> We observe a different line shape depending on the position of the LSPR of the isolated disk (red curve in each frame) with respect to  $\lambda_{\rm R}$  (solid green line in each frame). It is worth noting that despite the broad line width of the LSPRs of ferromagnetic nanoantennas we can induce sharp LSMs even if their peaks are spectrally far from the diffraction edge; this enables a large degree of freedom in tuning the optical and MO properties of the MPC without changing the lattice parameter. Given the symmetry of the MPC considered here, it is clear that the LSM-modulated Qext does not change when the array is illuminated by either x- or ypolarized radiation impinging perpendicularly on the array plane. If now the circular nanoantennas are replaced by elliptical shaped ones, the 90° rotational symmetry of the MPC is broken resulting in two different, spectrally separated LSPRs for x- and y-polarized incident radiation (or, equivalently, by rotating the sample by 90° while retaining the light polarization). As a result, two LSMs with different Fano line shapes are excited depending on the relative orientation of the elliptical nickel particles with respect to the polarization of the incident light, as sketched in the left panel of Figure 1b. Additionally, in the case of ferromagnetic nanoantennas, the intrinsic MOA activated by the application of a magnetic field perpendicular to the crystal plane leads to the simultaneous and coherent excitation of two in-plane orthogonal LSPRs, as demonstrated for randomly arranged ferromagnetic elliptical nanodisks.<sup>20</sup> The resonant diffractive coupling of these two antenna modes induced by the periodicity of the square lattice considered here will then lead to the simultaneous excitation of two perpendicular LSMs with different Fano resonant line shapes that can be precisely tailored through the design of the individual building blocks of the MPC.

In order to verify the physical picture presented above, we first designed and fabricated using e-beam lithography (EBL) a sample made of randomly distributed elliptical nickel antennas on a glass substrate. The elliptical nanoantennas have an inplane aspect ratio AR  $\approx$  2.5 to display LSPRs at markedly different wavelengths when the two axes are selectively excited by linearly polarized radiation impinging perpendicular to the sample surface. The in-plane dimensions of the antennas are 100 nm for the short axis (SA) and 250 nm for the long axis (LA). The thickness of the antennas is 30 nm. The system was then embedded in a homogeneous medium with n = 1.5 after immersion in refractive index matching oil. A SEM image of a portion of the fabricated system is shown in the top panel of Figure 2a. The Qext spectra of this sample for the two polarizations of the incident radiation are shown in the central panel of Figure 2b (dashed blue line and dashed red line for light polarization along SA- and LA-direction, respectively). The spectra show broad peaks at 580 nm (polarization along SA) and 1050 nm (polarization along LA) arising from the excitation of the corresponding LSPRs.

We then fabricated an MPC in which the same elliptical nanoantennas are arranged on a square array with a lattice constant a = 400 nm. We assume the LA and SA lying along the x- and y-direction as sketched in the left panel of Figure 1b. As for the random sample, the MPC was fabricated on a glass substrate and embedded in refractive index matching oil with n= 1.5. A SEM image of a portion of the fabricated system is shown in the bottom panel of Figure 2a. The experimental Q<sub>ext</sub> spectra of the MPC are plotted in the central panel of Figure 2b with solid lines using the same color code as for the random sample. When the electric field of the incident radiation oscillates along the LA-direction (red curve), we observe the excitation of a LSM with a Fano line shape featuring a dip around  $\lambda_{\rm R} = a \cdot n = 600$  nm, followed by a broad peak in the near-infrared region close to 1000 nm. In contrast, when the incident electric field is parallel to the SA-direction (blue curve), a LSM is excited with a dip around the same  $\lambda_{\rm R}$  but now followed by a narrow and sharp peak in the visible spectral region close to 650 nm. This behavior was expected based on the discussion of Figure 1 (see right panel of Figure 1b). Moreover, such a behavior is well reproduced and interpreted theoretically by means of the CDA, which allows for a compact analytical formulation of the Qext spectra of periodic arrays of nickel nanoantennas assuming that they radiate as coupled oscillating dipoles.<sup>35,37</sup> The  $Q_{ext}$  for the two polarizations of the incident field can be expressed as follows

$$Q_{\text{ext}-\text{LA}} \propto \text{Im}\{\tilde{\alpha}_{xx}^{\text{eff}}\} = \text{Im}\left\{\frac{1}{\frac{1}{\tilde{\alpha}_{xx}} - \varepsilon_0 \tilde{\varepsilon}_M \tilde{S}_{xx}^{\text{FF}}(\overline{y}_i)}\right\}$$
(1)

$$Q_{\text{ext-SA}} \propto \text{Im}\{\tilde{\alpha}_{yy}^{\text{eff}}\} = \text{Im}\left\{\frac{1}{\frac{1}{\tilde{\alpha}_{yy}} - \varepsilon_0 \tilde{\epsilon}_M \tilde{S}_{yy}^{\text{FF}}(\overline{x}_i)}\right\}$$
(2)

where  $\tilde{\alpha}_{xx}$  ( $\tilde{\alpha}_{yy}$ ) is the polarizability tensor elements of the individual dipole along the LA- (SA-) direction,  $\tilde{e}_{\rm M}$  is the permittivity of the embedding medium, and  $\tilde{S}_{xx}^{\rm FF}(\hat{y})$  ( $\tilde{S}_{yy}^{\rm FF}(\hat{x})$ ) is the dipolar far-field interaction factor for LA (SA) illumination (see Supporting Information). The last term depends only on the geometrical parameters of the lattice, and specifically on the y- (or x-) direction, indicated by the  $\hat{y}$  (or  $\hat{x}$ ) unit vector dependence. From the experiments and the above formulas it is straightforward to see that the Qext spectrum of the MPC, when the incident field is oscillating along the LA-direction, is determined by a LSM involving the antennas LA-LSPR and the far-field diffractive interaction along the SA-direction (as expressed by the y-direction dependence of  $\tilde{S}_{xx}^{FF}(\hat{y})$  and demonstrated in Supporting Information, Figure S1 and related discussion). Likewise, the Qext spectrum for light polarized along the SA-direction is determined by a LSM resulting from the SA-LSPR excited in each antenna and the far-field diffractive interaction along the LA-direction. In the present case of square lattices  $\tilde{S}_{xx}^{FF}(\tilde{y}) = \tilde{S}_{yy}^{FF}(\hat{x})$ , so  $\lambda_R$  does not change and the line shape of the LSMs is determined by the relative position between the involved LSPR peak and the diffraction edge. The fact that the LSMs are determined by the far-field term of the dipole-dipole interaction along the direction perpendicular to the induced electric dipole in each nanoantenna has indeed been observed experimentally in MPCs with cylindrical magnetic nanoantennas on a rectangular lattice.45 Two important observations can be made at this point regarding the optical properties of a MPC made of ferromagnetic nanoantennas: (i) the periodic arrangement induces a marked sharpening of the spectral features near  $\lambda_{\rm R}$ , as compared to the  $Q_{\rm ext}$  of randomly distributed antennas<sup>48</sup> and, more importantly, (ii) the optical anisotropy  $\Delta Q$  induced in the system, defined as  $Q_{\rm ext-LA} - Q_{\rm ext-SA}$  (violet solid curve in the right panel of Figure 2b), shows a clear enhancement close to the diffraction edge if compared to the randomly distributed nanoantennas case (dashed violet curve). The second effect, which can be seen in previously reported studies on PCs of anisotropic gold nanoantennas,<sup>49</sup> shows how the effects of the shape anisotropy of the individual building block on the optical properties of the system can be amplified and tuned via LSMs design.

We then focused our attention on the MOA of our MPC, disclosing the most interesting effects arising from the application of an external magnetic field, as sketched in the left panel of Figure 2c. When the incident electric field oscillates along the SA-(LA-)direction and a magnetic field is applied perpendicularly to the crystal plane, the magnetic field activated SOC inherent to each individual nanoantenna induces a second oscillating dipole in the LA-(SA-)direction with a resonant behavior according to the LA-(SA-)LSPR of the nanoantennas. The MO response of the MPC is determined both by the electric field scattered by the SOC-induced dipole along the LA-direction and by that originating from the dipole induced directly by the electric field of the incident radiation along the SA-direction. Because of the phase lag between these two oscillating electric dipoles, the magnetic field induced change in the polarization of the reflected (transmitted) light is conveniently defined through the following complex Kerr (Faraday) angles for each incidence radiation (LA or SA)

$$\frac{\tilde{E}_{SA}}{\tilde{E}_{LA}} = \tilde{\Phi}_{LA} = \theta_{LA} + i\varepsilon_{LA}$$
(3)

$$\frac{\tilde{E}_{LA}}{\tilde{E}_{SA}} = \tilde{\Phi}_{SA} = \theta_{SA} + i\varepsilon_{SA} \tag{4}$$

where the real ( $\theta$ ) and imaginary ( $\varepsilon$ ) parts are the MO-induced rotation (with respect to the incident polarization plane) and ellipticity of the outward reflected (transmitted) electric field, respectively. Physically, the MOA is defined as the modulus of the complex Kerr angle, namely  $|\tilde{\Phi}| = \sqrt{\theta^2 + \varepsilon^2}$ .<sup>33</sup> Following the same reasoning used for explaining the Qext spectra in Figure 2b, we now expect the MOA to be determined by MO-LSMs stemming from diffractive coupling of the electric fields emitted by the SOC-induced dipoles and acting in the direction perpendicular to them, namely parallel to the incident radiation polarization, as sketched in the left panel of Figure 2c. After saturating the magnetization of the nanoantennas by applying a magnetic field  $|H_{sat}| = 3$  kOe perpendicular to the MPC plane, the MOA of our MPC was measured as function of the wavelength of the incident light in reflection (Kerr) geometry (see Supporting Information for details on the experimental set-up used). The experimental MOA spectra are shown in the central panel of Figure 2c, where the red and blue curves refer to an incident electric field oscillating along the LA- and SAdirection, respectively. It is worth noting how the MOA features notably resemble those of the Qext measured with the polarization of the incident field along the perpendicular direction. Similar, but configurationally different effects between optical and magneto-optical spectra have also been

observed in MPCs made of rectangular arrays of circular nickel nanodisks.<sup>45</sup> The experimental results in the central panel of Figure 2c confirm the intuitive picture shown in the cartoon in the left panel and are well reproduced by the following analytical formulas governing the Kerr (Faraday) complex angles for LA- and SA-direction (see Supporting Information for the complete derivation)

$$\begin{split} \tilde{\Phi}_{\text{LA}} &\approx \frac{\tilde{p}_{y}}{\tilde{p}_{x}} = \frac{\tilde{\alpha}_{yx}}{\tilde{\alpha}_{xx}} \left( \frac{1}{1 - \varepsilon_{0} \tilde{\epsilon}_{\text{M}} \tilde{\alpha}_{yy} \tilde{S}_{yy}} \right) = \frac{\tilde{\alpha}_{yx}}{\tilde{\alpha}_{xx} \tilde{\alpha}_{yy}} \left( \frac{1}{\frac{1}{\tilde{\alpha}_{yy}} - \varepsilon_{0} \tilde{\epsilon}_{\text{M}} \tilde{S}_{\text{FF}}^{\text{FF}}(\bar{x}_{i})} \right) \end{split}$$
(5)  
$$\tilde{\Phi}_{\text{SA}} &\approx \frac{\tilde{p}_{x}}{\tilde{p}_{y}} = \frac{\tilde{\alpha}_{xy}}{\tilde{\alpha}_{yy}} \left( \frac{1}{1 - \varepsilon_{0} \tilde{\epsilon}_{\text{M}} \tilde{\alpha}_{xx} \tilde{S}_{xx}} \right) = \frac{\tilde{\alpha}_{xy}}{\tilde{\alpha}_{xx} \tilde{\alpha}_{yy}} \left( \frac{1}{\frac{1}{\tilde{\alpha}_{xx}} - \varepsilon_{0} \tilde{\epsilon}_{\text{M}} \tilde{S}_{xx}^{\text{FF}}(\bar{y})} \right)$$
(6)

where  $\tilde{p}_x$  and  $\tilde{p}_y$  are the LA- and SA-components of the dipole moment of the individual nickel particle and  $\tilde{\alpha}_{yx}$ ,  $\tilde{\alpha}_{xy}$  are the offdiagonal elements of the polarizability tensor. Because the MO response is much smaller than the optical one,  $\tilde{\alpha}_{yx} = -\tilde{\alpha}_{xy} \approx$  $\tilde{x}_{\text{SOC}}\tilde{\alpha}_{xx}\tilde{\alpha}_{yy}$  with  $\tilde{x}_{\text{SOC}} = -iQ\tilde{\varepsilon}_{\text{Ni}}\tilde{\varepsilon}_M/[V(\tilde{\varepsilon}_{\text{Ni}} - \tilde{\varepsilon}_M)^2]$  and eqs 5 and 6 can be simplified as follows<sup>50</sup>

$$\tilde{\Phi}_{\rm LA} \approx \frac{-iQ\tilde{\epsilon}_{\rm Ni}\tilde{\epsilon}_{\rm M}}{V(\tilde{\epsilon}_{\rm Ni} - \tilde{\epsilon}_{\rm M})^2} \left(\frac{1}{\frac{1}{\tilde{a}_{yy}} - \epsilon_0 \tilde{\epsilon}_{\rm M} \tilde{\delta}_{yy}^{\rm FF}(\overline{x}_i)}\right)$$
(7)

$$\tilde{\Phi}_{\rm SA} \approx \frac{i Q \tilde{\varepsilon}_{\rm Ni} \tilde{\epsilon}_{\rm M}}{V (\tilde{\epsilon}_{\rm Ni} - \tilde{\epsilon}_{\rm M})^2} \left( \frac{1}{\frac{1}{\tilde{\alpha}_{xx}} - \varepsilon_0 \tilde{\epsilon}_{\rm M} \tilde{S}_{xx}^{\rm FF}(\overline{y}_i)} \right)$$
(8)

where Q is the Voigt constant expressing the strength of the SOC,  $\tilde{\varepsilon}_{Ni}$  is the dielectric tensor of bulk nickel and V the volume of the nanoparticle. From eqs 7 and 8 we note that the Kerr (Faraday) complex angles  $ilde{\Phi}_{LA}$  and  $ilde{\Phi}_{SA}$  depend only on the magneto-optical induced response ( $\tilde{\alpha}_{vv}$  in  $\tilde{\Phi}_{LA}$  and  $\tilde{\alpha}_{xx}$  in  $ilde{\Phi}_{ ext{SA}}$ ), in analogy with what already has been demonstrated for noninteracting magnetoplasmonic nanoantennas.<sup>20</sup> A qualitative but transparent physical insight of the result summarized in eqs 7 and 8 can be gained as follows. The incident radiation excites a LSM, which determines the optical response of the array. This primary LSM drives the second MO-LSM through the inherent SOC, and therefore the magneto-optical response contains both LSMs, the primary optical LSM and the SOCinduced MO-LSM. However, the MOA, which is the quantity that identifies the polarization of the reflected (transmitted) radiation, is the magneto-optical response divided by the optical one. It is this normalization that "simplifies out" from the MOA any effect arising from the primary LSM that is directly induced by the incident radiation. What emerges from the experimental results, which are excellently reproduced by eqs 1-8 given above (see also Supporting Information, Figure S1), is that the MOA of the MPC studied here is controlled by the following two key elements: (i) the SOC-induced polarizability of the individual dipolewhich is activated by the application of a magnetic field and it is transverse to that directly excited by the electric field of the incident light and (ii) the diffractive coupling along the direction parallel to that of the linear polarized incident electric field leading to the excitation of a MO-LSM with a Fano spectral line shape. Remarkably, we observe that the excitation of MO-LSMs for both polarizations of the incident field produces a marked spectral reshaping of the MOA (see solid lines in the central panel of Figure 2c), which are dependent on the relative orientation of the elliptical nickel particles with respect to the polarization of the incident light. Moreover, these noteworthy effects result in a strong enhancement of the MOA of the MPC when compared to the random sample (see solid and dashed lines in central panel of Figure 2c), thus opening more versatile opportunities for fine engineering of the MO response of the MPC. Finally, we also looked at what happens up on reducing the strength of the static magnetic field IHI: while the spectral shape of the MOA does not change, its intensity progressively reduces as IHI is reduced from  $|H_{sat}|$  (see Supporting Information and Figure S2). This dependence of the MOA on the strength of the magnetic field IHI, opens a clear path to the active and remote tuning of the MOA.

To better appreciate and quantitatively understand the advantage of exciting LSMs in MPCs with respect to random systems, we would now like to focus the reader's attention on the quantity plotted in the right panel of Figure 2c for the case studied here. In analogy with  $\Delta Q$ , we can define MOA anisotropy as

$$\Delta_{\text{MOA}} = |\tilde{\Phi}_{\text{LA}}| - |\tilde{\Phi}_{\text{SA}}| \tag{9}$$

Primarily, the intrinsic shape anisotropy in the randomly distributed nanoantennas is also expected to induce a  $\Delta_{MOA}$  in analogy with the  $\Delta Q$  in the optical response. However, in the disordered system this intrinsic anisotropy leads to much weaker effects in the MO spectra (violet dashed lines in the  $\Delta_{MOA}$  spectra in the right panel of Figures 2c). On the other hand, in the MPC the intrinsic shape anisotropy of the individual nanoantenna is largely amplified by the coherent farfield diffractive coupling between the nanoantennas. Indeed, the striking result observed in the right panel of Figure 2c is that the MPC displays a significant difference between the LA- and the SA-aligned MOAs (violet solid curve in the  $\Delta_{MOA}$  spectra). For instance, where we see almost zero  $\Delta_{MOA}$  for the randomly distributed nanoantennas at ~670 nm, the MPC shows the maximum of  $\Delta_{\text{MOA}}$ . All the experimental results shown here are in excellent agreement with the results from finite-difference time-domain (FDTD) and CDA techniques, as thoroughly discussed in the Supporting Information.

To delve more deeply the physics disclosed above and to check how far one can control the spectral line shape of the MOA of a MPC through the in-plane plasmonic anisotropy tuning, we performed CDA calculations by fixing the square array pitch and varying the aspect ratio of the nanoantennas. We shifted the spectral position of one LSPR at a time along the principal axes of the ellipses with respect to  $\lambda_{R}$ . We first simulated square arrays of nickel nanodisks with SA = 150 nm, a thickness equal to 30 nm and a LA size ranging from 187.5 to 487.5 nm (i.e., AR 1.25-3.25, respectively, see the cartoon in the top panel of Figure 3a). The square lattice parameter was fixed to 500 nm; the reason for this change was to explore a wider range of LSPR blue-shifts with respect to  $\lambda_{R}$ , given that our experimentally accessible lower wavelengths range was limited to 500 nm. A homogeneous medium with refractive index n = 1.5 was assumed to model the experimental matching-oil condition. Therefore, the Rayleigh's anomaly condition is in this case  $\lambda_{\rm R} = a \cdot n = 750$  nm. In the bottomleft panel of Figure 3a, we show the MOA for incident electric field oscillating along both the SA- (solid curves) and LA-(dashed curves) direction. In the latter case, the MO-LSM is controlled by the lattice parameter along the LA-direction and



**Figure 3.** (a) Top panel: LSPRs of an isolated elliptical nanoantenna. The SA diameter is 150 nm. The LA diameter is changed from 187.5 nm to 487.5 nm (AR 1.25–3.25). The pitch a is 500 nm along both LA- and SA-direction and the thickness t of each antenna is 30 nm. (a) Left-bottom panel: MOA for  $E_{inc}$  parallel to the LA of the antennas (dashed curves) and for *E* parallel to the SA of the antennas (solid curves). (a) Right-bottom panel:  $\Delta_{MOA}$  of the MPC for different AR ranging from 1.25 to 3.25. (b) Top panel: as in (a), but with the LA diameter equal to 150 nm. The SA diameter is varied from 75 nm to 50 nm; a = 500 nm along both LA- and SA-direction and t = 30 nm. (b) Left-bottom panel: MOA for  $E_{inc}$  parallel to the LA of the antennas (solid curves) and for  $E_{inc}$  parallel to the SA of the antennas (dashed curves). (b) Right-bottom panel:  $\Delta_{MOA}$  for the MPC for different AR ranging from 2 to 3. All the data are calculated using CDA.

the SA size of the nickel nanoantennas (MO-SA-LSPR), which are both fixed. As a result, we observe the MOA intensity slightly decreasing when passing from AR = 1.25 to AR = 3.25without too much spectral change (dashed lines). In striking contrast, when the incident electric field oscillates along the nanoantennas SA-direction (solid curves), we observe clear changes in the MOA spectral line shape, as in this case the MO-LSM is controlled through the nanoantennas MO-LA-LSPR, which is red shifting as we increase the AR. These effects can be used to enhance and tune the MOA of a MPC by designing the shape anisotropy of the individual building block, which is



**Figure 4.** Top panel: elliptical antennas with in-plane dimensions of 150 nm and 400 nm, and thickness t = 30 nm. (a) Top: SEM image of a portion of the ordered array; pitch a = 500 nm; scale bar 500 nm. (a) Bottom: SEM image of a portion of the random array; scale bar 500 nm. (b) Qext (top) and MOA (bottom) of the periodic (solid lines) and random (dashed lines) arrays, for  $E_{inc}$  applied along the LA (red lines) and along the SA (blue lines). (c)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the MPC (violet solid lines) and random array (violet dashed lines). Bottom panel: elliptical antennas with in-plane dimensions of 80 nm (SA) and 150 nm (LA), and thickness of 30 nm. (d) Top: SEM image of a portion of the ordered array; a = 500 nm. (d) Bottom: SEM image of a portion of the random array. Scale bars 500 nm. (e)  $Q_{ext}$  (top) and MOA (bottom) of the periodic (continuous lines) and random (dashed lines) arrays, for  $E_{inc}$  applied along the SA (blue lines). (f)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the MPC (violet dashed lines) and along the SA (blue lines). (f)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the array (violet dashed lines) and along the SA (blue lines). (f)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the MPC (violet dashed lines) and along the SA (blue lines). (f)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the MPC (violet dashed lines) and along the SA (blue lines). (f)  $\Delta Q$  (top) and  $\Delta_{MOA}$  (bottom) for the MPC (violet solid lines) and random array (violet dashed lines).

reinforced through the collective behavior produced by the coherent excitation of the lattice modes radiated to the far-field. We plot also  $\Delta_{MOA}$  in the bottom-right panel of Figure 3a. Indeed, we observe that by manipulating the MO-LSMs line shapes through varying the AR, we can produce a significant variation and enhancement of  $\Delta_{MOA}$  in a narrow spectral region centered around 850 nm, which is clearly controlled by the LA of the nanoantennas. Here the peak position increases with increasing AR and saturates for AR = 2.75.

Complementarily, in Figure 3b we analyze the effects of SA-LSPR tuning on the MO-LSMs spectral line shapes (see the cartoon in the top panel of Figure 3b). We simulated square MPCs made of nickel nanodisks with LA = 150 nm, t = 30 nm and SA size ranging from 75 to 50 nm (i.e., AR 2–3, respectively). As in the previous case, a = 500 nm and n = 1.5. The results of the CDA calculations, displayed in the bottom panel of Figure 3b, show that now the huge variation in the MOA spectral line shape and intensity is occurring when the

incident electric field is polarized along the LA-direction (solid curves in the bottom-left panel of Figure 3b). This is consistent with the physical picture developed so far in this Letter and summarized by eq 5, because in this case the MOA depends on the MO-LSM tuning caused by the MO-SA-LSPR blue shift with respect to  $\lambda_{\rm R}$  induced and controlled by varying the SA of the elliptical nanoantennas. In a straightforward analogy with the previous case, for incident radiation polarized along the SAdirection the MOA spectral line shape shows only minor changes, because the MO-LSM in this case is controlled by the nanoantenna MO-LA-LSPR, which now is the fixed one (see dashed curves in the bottom-left panel of Figure 3b). For the case analyzed in Figure 3b, a remarkable property can be bestowed on the MPC through the MO-LSM tuning by varying the AR. In this case a complete isotropic MOA in a specific and narrow spectral region is observed despite the anisotropic character of the individual dipoles (see the bottom-right panel of Figure 3b). This interesting effect, which is caused by the ordered nature of the system, can be achieved for an AR between 2 and 2.25 for the nanoantennas size and lattice parameter ranges considered here. Overall, the clear outcome of the modeling efforts reported in Figure 3 is that MO-LSMs line shapes tuning in MPCs, achievable through a precise control of the size and shape of the constituent nanoantennas with respect to the lattice parameter, offers a tremendous potential for designing MPCs with a preselected and finely tailored MO response. Therefore, it is worth mentioning that all the abovediscussed effects arising from the excitation of LSMs in MPC, can be transferred to any other spectral position by varying the lattice parameter and/or the overall sizes of the nanoantennas principal axes.

To experimentally prove the versatile potentiality of MO-LSMs tuning in MPCs outlined above, we fabricated two MPCs aimed at achieving the two key cases presented in Figure 3, namely the optimization of  $\Delta_{MOA}$  as compared with the case presented in Figure 2, and the achievement of MOA isotropy in selected and narrow spectral ranges. Control samples with randomly arranged nanoantennas with the same filling factor and dimensions of the individual units as in the corresponding MPCs were fabricated and measured as reference. To prove the first case, that is, the amplification of  $\Delta_{MOA}$  in a selected spectral range, the in-plane dimensions of the elliptical nanoantennas were chosen to be 150 and 400 nm (SA and LA, respectively). The thickness of the nanoantennas is 30 nm and the lattice parameter of the MPC is 500 nm. Because in this MPC the LA dimension of the nanoantennas is not much smaller than the wavelength of the incoming light, we checked its optical response with both CDA and FDTD simulations. As discussed in the Supporting Information and shown in Figure S3, we found that the dominant mode has a dipolar character also for nickel nanoantennas with LA = 400 nm. We can therefore conclude that the formation of LSMs is always dominated by the strongly radiating dipolar mode of the nanoantennas, even in MPC made of nickel nanoantennas of large size. This conclusion is further corroborated by the excellent agreement between the spectra calculated using the CDA (Figure 3) and those measured experimentally (Figure 4). It is worth mentioning that for noble metal nanoantennas of similar size, higher order modes would be indeed relevant.<sup>51</sup> Figure 4a shows the SEM image of a portion of the MPC (top panel) and the randomly distributed nanoantennas (bottom panel) fabricated by EBL. The geometrical parameters of this first system are closed (AR = 2.67) to those of the MPC

predicted to display the maximum of  $\Delta_{MOA}$  (AR = 2.75), studied in Figure 3a. In the top panel of Figure 4b, we show the  $Q_{\text{ext}}$  spectra of both the MPC and the randomly distributed nanoantennas. For the MPC, we observe two very different sharp features around the diffraction edge at 750 nm for  $E_{inc}$ applied along the LA (red solid curve) and the SA (blue solid curve), while for the random system we see two spectrally displaced and very broad plasmonic peaks (red and blue dashed curves). Similarly, both the MOA<sub>LA</sub> and MOA<sub>SA</sub> spectra, which are shown in the bottom panel of Figure 4b, feature the expected Fano line shape with a dip at 750 nm and enhanced peaks at around 850 nm (light polarization along LA, solid red line) and around 1100 nm (light polarization along SA, solid blue line). This behavior is in excellent agreement with the calculations presented in Figure 3a and leads to the predicted peak of  $\Delta_{MOA}$  at a wavelength of about 825 nm. Apart from confirming the theoretical predictions, a key aspect conveyed by these experiments is the outstanding enhancement of  $\Delta_{MOA}$ produced by the resonant excitation of MO-LSMs in the MPC when compared to that produced in a disordered assembly of the same constituent units (same filling factor for the MPC and the random sample). This is clearly appreciable in the bottom panel of Figure 4b, where the MOA<sub>LA</sub> and MOA<sub>SA</sub> spectra of the random control sample are shown for direct comparison (red and blue dashed curves, respectively). The advantage of exciting LSMs is even more clearly visible in Figure 4c, where we plot the  $\Delta Q$  (top panel), and  $\Delta_{MOA}$  (bottom panel) for both the MPC (violet solid lines) and the random system (violet dashed lines). In this case we achieved a  $\Delta_{MOA}$  that is a factor of 2 greater than that shown in Figure 2c.

Finally, in order to experimentally prove the second case of interest predicted by the CDA calculations, namely the achievement of a wavelength-dependent isotropy of the MOA in a selected and narrow spectral range, we fabricated a MPC composed of 30 nm thick elliptical nanodisks with in-plane axes of 150 nm (LA-direction) and 80 nm (SA-direction) arranged in the same 500 nm pitch square lattice. The AR of the ellipses were chosen to be as close as possible, compatibly with our nanofabrication limits, to those prescribed by simulations in order to achieve the desired effect (AR = 1.87 for the real sample versus AR between 2 and 2.25 for the simulations). Also for this case, we fabricated a control random sample with the same type of nanoantennas and identical filling factor as for the MPC. SEM images of a portion of both samples are shown in Figure 4d. The experimental  $Q_{\text{ext}}$  and MOA spectra of the MPC are shown in Figure 4e (top and bottom panels, respectively). The MOA<sub>LA</sub> and the MOA<sub>SA</sub> spectra (red and blue solid curves in bottom panel of Figure 4e, respectively) reproduce very well the line shapes predicted by calculations and reported in the bottom-left panel of Figure 3b, and confirm that also in this case the excitation of MO-LSMs in the MPC induces profound differences in the MOA spectra when compared to a disordered assembly of the same constituent units (red and blue dashed lines in the bottom panel of Figure 4e). Most importantly,  $\Delta_{\rm MOA}$  spectrum plotted in the bottom panel of Figure 4f for the MPC case (violet solid curve) excellently matches the modeling predictions as it displays the achievement of an almost perfect MOA isotropy in a narrow range of wavelengths around 770 nm. One final striking result is that the MOA<sub>LA</sub> and the MOA<sub>SA</sub> spectra for the random sample (dashed red and blue curves in Figure 4e) as well as  $\Delta_{MOA}$  (violet dashed curve in the bottom panel of Figure 4f) indicate that this sample shows a negligible  $\Delta_{\mathrm{MOA}}$  over the whole spectral range explored. This further

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confirms the key role played by the excitation of MO-LSMs in determining the MOA of a MPC and the huge potential of using ordered systems to increase MO effects and magnetoplasmonic metasurfaces.

Before concluding, it is worth briefly addressing the effects arising from varying the lattice symmetry. The use of nonrectangular arrangements, like hexagonal or honeycomb lattices, is not expected to display additional effects different from those occurring in square lattices, as reported by Humphrey and Barnes.<sup>43</sup> Indeed, as discussed in ref 43, such symmetries are highly isotropic in terms of LSMs excitation (see Figure 7 of the ref 43.). In contrast, rectangular lattices provide an additional source of anisotropy<sup>45</sup> that can be used in combination with the anisotropy of the individual building block to induce a compensation or even a change of sign in the anisotropy of Qext and of the MOA at selected wavelengths. This is indeed the case, as shown in Figure S4, where the exemplary case of the MPC treated in Figure 4a-c is chosen. Figure S4 shows that by choosing two different lattice parameters along the antennas principal directions one can induce either enhancement or suppression of  $\Delta Q$  and  $\Delta_{MQA}$ .

In summary, we presented a new concept of magnetoplasmonic crystal based on the excitation of Fano lattice surface modes in periodically arranged magnetic antennas that show anisotropic in-plane plasmonic responses. Our findings reveal that the excitation of lattice surface modes in this kind of system is governed by the polarizability of the single antenna and the relative position between localized resonances in the individual building blocks and diffraction edges induced by the periodicity of the crystal lattice. The different in-plane optical response along two transverse directions, which are coupled by the application of an external magnetic field, leads to a significant enhancement and a fine-tuning of both optical and magneto-optical effects in either narrow or large regions of the visible and near-infrared spectrum. In particular, the strengthening of magneto-optical effects by the coherent excitation of radiative far-field interactions between the nickel particles shows up the great potential rising from the excitation of lattice surface modes in magnetoplasmonic metasurfaces based on magnetic nanonatennas, opening up excellent nanoengineering opportunities towards enhanced and generally designed magneto-optical properties. We envisage the concept of the magnetoplasmonic crystal presented here will enable novel metamaterials or applications, such as nanostructured magnetically-tunable nonreciprocal optical isolators or notch-filters for the control of light polarization states, as well as improved nanoscale magnetoplasmonic refractometric and molecularlevel detection schemes.

## ASSOCIATED CONTENT

#### **S** Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nano-lett.6b00084.

Samples fabrication; optical and magneto-optical characterization; single particle in the dipole approximation; coupled dipole approximation (CDA); optical and magneto-optical properties with CDA; finite-difference time-domain (FDTD) simulations; size tweaking for the nanoantenna modeling for the case shown in Figure 2; far-field coupling in periodic arrays for the case shown in Figure 2; magneto-optical activity at different strengths of the applied magnetic field; localized resonances for large nickel particles; lattice symmetry effects in rectangular arrays. (PDF)

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#### **Author Contributions**

N.M., L.B., and M.P. contributed equally.

N.M. and P.V. conceived the idea and performed optical and magneto-optical characterization. L.B., M.K.S., N.Z., and J.A. performed theoretical modeling, CDA calculations, and FDTD simulations. M.P. fabricated the samples and helped with CDA calculations. N.M., L.B., and P.V. discussed the results and wrote the manuscript with the input from all the authors.

# Notes

The authors declare no competing financial interest.

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