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Published in:
Optics Express

DOI:
10.1364/OE.24.003562

Published: 22/02/2016

Document Version
Publisher's PDF, also known as Version of record

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Please cite the original version:
Kataja, V., Pourjamal, S., & Van Dijken, S. (2016). Magnetic circular dichroism of non-local surface lattice resonances in magnetic nanoparticle arrays. Optics Express, 24(4), 3562-3571. https://doi.org/10.1364/OE.24.003562
Magnetic circular dichroism of non-local surface lattice resonances in magnetic nanoparticle arrays

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Abstract: Subwavelength metallic particles support plasmon resonances that allow extreme confinement of light down to the nanoscale. Irradiation with left- and right hand circularly polarized light results in the excitation of circular plasmon modes with opposite helicity. The Lorenz force lifts the degeneracy of the two modes in magnetic nanoparticles. Consequently, the confinement and frequency of localized surface plasmon resonances can be tuned by an external magnetic field. In this paper, we experimentally demonstrate this effect for nickel nanoparticles using magnetic circular dichroism (MCD). Besides, we show that non-local surface lattice resonances in periodic arrays of the same nanoparticles significantly enhance the MCD signal. A numerical model based on the modified long wavelength approximation is used to reproduce the main features in the experimental spectra and provide design rules for large MCD effects in sensing applications.

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OCIS codes: (210.3810) Magneto-optic systems; (160.5298) Photonic crystals; (250.5403) Plasmonics.

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1. Introduction

Surface plasmon resonances in metallic nanoparticles give rise to optical properties that are not available in naturally occurring materials. The ability of surface plasmons to confine light to subwavelength dimensions is useful for several applications including label-free biosensors and optical communication technologies [1]. Simple spherical and linear noble metal antennas...
can be reshaped to support ring currents and create negative-refractive-index materials [2] while spatial variations of the antenna shape can be used to control the phase of scattered radiation [3]. Phase-gradient metasurfaces display a range of novel phenomena such as selective scattering of left- and right hand circularly polarized light. This effect, however, is inherent to the pre-defined geometry of the antennas and, hence, cannot be tuned actively during device operation.

On the other hand, the interaction of light with magnetic materials gives rise to a variety of magneto-optical phenomena such as the Faraday and Kerr effects. The magnitude of these effects can be altered reversibly by control of the magnetization direction using external magnetic fields. Recently, magneto-plasmonic systems combining magneto-optical effects and enhancements of electric fields in subwavelength nanostructures have attracted considerable interest [4]. For example, magneto-plasmonics has been studied for noble metal plasmonic crystals on top of magneto-optically active dielectric garnets [5–9], near-field coupled noble-metal/ferromagnetic sandwiches [10,11] and dimers [12], and pure magnetic nanostructures that support localized surface plasmon resonances (LSPRs) [13–16]. In addition, efficient coupling of LSPRs in magnetic nanoparticles to the diffracted orders of a periodic lattice, leading to non-local surface lattice resonances (SLRs) with narrow and asymmetrical spectral lineshapes, has recently been demonstrated [17]. Approaches involving pure magnetic nanoparticles are relatively straightforward and allow for direct assessments of interactions between magnetization, magneto-optical activity, and local and non-local plasmon modes.

Faraday and Kerr effects are commonly used to probe the magneto-optical activity of ferromagnets. Both measurement concepts utilize linearly polarized light with zero spin angular momentum (SAM). Here, we report on the interaction between light with non-zero SAM and ferromagnetic nanoparticles using magnetic circular dichroism (MCD) spectroscopy. Two types of samples are studied; random nickel nanoparticles with perpendicular magnetization (to probe the interaction between LSPRs and light with opposite helicity) and periodic nickel nanoparticle arrays (to analyze far-field diffractive coupling between circular plasmon modes). The main features of the experimental MCD spectra are reproduced by an analytical model based on the coupled dipole approximation.

2. Experimental

Cylindrical nickel nanoparticles with a diameter of 120 nm and a thickness of 30 nm were fabricated on glass substrates using electron beam lithography (EBL). Four different samples were measured: one with a random distribution of nanoparticles and three with periodic arrangements of nanoparticles in square arrays. The size of the arrays was 500 × 500 μm and the periodicity of the particles was varied (p = 400, 450, and 500 nm) to study the effect of SLRs on the MCD spectra. The samples were immersed in index matching oil (Cargille Laser Liquid, n = 1.523) to provide a symmetric refractive index environment.

The optical properties of the samples were characterized in transmission geometry with normal incident light. The wavelength of the light (λ) was varied from 500 nm to 1100 nm using a broadband laser and an acousto-optical filter (NKT Photonics SuperK EXW-12). The intensity of transmitted light was measured with a photodetector and transmittance spectra were obtained by normalizing the measured intensity to the laser intensity. To enable a straightforward comparison of the experimental data with model calculations presented later, we used extinction spectra (E = 1 − T) to characterize the optical properties of the samples.

MCD was measured with a lock-in technique based on photoelastic modulation (Hinds Instruments PEM I/F50) of the incoming light between left- and right hand circularly polarized states. The difference between optical transmittance (T) for opposite helicities was directly measured by phase-sensitive detection at the modulation frequency. An out-of-plane magnetic field of 150 mT was applied during the measurements to ensure fully saturated perpendicular magnetization in the nickel nanoparticles. MCD spectra were recorded for
saturating positive and negative field directions and both measurements were subsequently subtracted to isolate dichroism arising from pure magnetic effects. The MCD signal in our experiments corresponds to 

$$\text{MCD} = \frac{(E_+ - E_-)/E}{(E_+ + E_-)/E},$$

with $E_+$ and $E_-$ the optical extinction of left- and right hand circularly polarized light. Scanning electron microscopy (SEM) images of two samples and a schematic illustration of the MCD measurement setup are shown in Fig. 1.

![SEM images of samples](image)

**Fig. 1.** SEM images of a sample with (a) a random distribution of nickel nanoparticles and (b) a square array of nickel nanoparticles with a periodicity of $p = 400$ nm. The imaged area is 3 x 3 μm. (c) Schematic illustration of the MCD measurement setup. In the experiments, a laser spot with a diameter of 300 μm and an out-of-plane magnetic field of 150 mT were used.

### 3. Optical extinction and MCD measurements

In the experimental configuration, left- and right hand circularly polarized light excite circular plasmon modes in the nickel nanoparticles. These modes are characterized by collective oscillations of conduction electrons in circular orbits. The direction of electron motion is perpendicular to the momentary electric field of the incoming light. For nickel nanoparticles with perpendicular magnetization, an additional Lorenz force acts on the electrons. The resulting forces are sketched in Fig. 2(a). Depending on the helicity of the incident light, the Lorenz force either adds to or subtracts from the centripetal force that is exerted by the electric field, which alters the confinement of electrons [18–20]. Therefore, the degeneracy of circular plasmon modes is lifted by the perpendicular magnetization of the nickel nanoparticles. The helicity-induced plasmon resonances are spectrally separated with the more confined mode occurring at higher frequency (shorter wavelength) in optical extinction spectra in Fig. 2(b). The difference between the extinction profiles corresponds to the MCD signal.
Fig. 2. (a) Sketch of forces acting on electrons in circular motion, i.e. when a LSPR is excited in a magnetic nanoparticle by circular polarized light. The electric field of the incident light gives rise to a force \( F_E \) and the perpendicular component of the magnetization acts on the circularly oscillating electrons via the Lorentz force \( F_B = e \mathbf{v} \times \mathbf{B} \). Depending on the helicity of the incident light the Lorenz force either adds to or opposes \( F_E \). (b) Extinction profiles of the non-degenerate circular plasmon modes. The difference between the extinction spectra determines the MCD signal. The MCD signal is zero at the LSPR resonance condition for non-magnetic nanoparticles (\( \omega_0 \)).

Experimental extinction \((1 - T)\) and MCD spectra of the sample with a random distribution of nickel nanoparticles are depicted in Fig. 3(a). A LSPR peak is visible at 700 nm in the extinction spectrum. Because of relatively high ohmic losses in nickel, the plasmon resonance is broad compared to that of noble metal nanoparticles. The lineshape of the MCD spectrum corresponds to the subtraction of two shifted extinction curves for left- and right hand circularly polarized light. As expected, the MCD signal reverses sign at the plasmon resonance condition, i.e. when the extinction of light by the nickel nanoparticles is maximum. From the asymmetrical shape of the MCD spectrum one can conclude that MCD becomes stronger towards infrared wavelengths. This effect is caused by an increase of the magneto-optical coupling strength (Voigt parameter \( Q \)) in the nickel nanoparticles at large wavelength [21]. The influence of \( Q(\lambda) \) on MCD spectra will be discussed in more detail in the next section.

Periodic arrangements of nanoparticles can give rise to collective plasmon excitations known as SLRs. These non-local resonances arise when LSPRs in metal particles of a periodic array couple to the diffracted orders of the lattice. For normal incident light, the diffracted orders of the lattice are given by \((a^2 + b^2)^{0.5} \lambda = np\), where \( n \) is the refractive index of the medium surrounding the particles, \( p \) is the periodicity of the lattice, and \( a \) and \( b \) are integers indicating the order of diffraction along the \( x \)- and \( y \)-axis of the array. In periodic arrays of noble metal nanoparticles, SLRs are manifested by narrow and intense optical resonances [22,23]. Similar effects and SLRs in magneto-optical spectra have been obtained for square and rectangular nickel nanoparticle arrays using linearly polarized light [17].

In the extinction spectra of Fig. 3(b), the minima at \( \lambda = 609, 685, \) and 762 nm correspond to the \( < + 1,0> \) and \( < -1,0> \) diffracted orders of the lattice with \( p = 400 \) nm, 450 nm, and 500 nm, respectively. In addition, a minimum associated with the diagonal \( <1,1> \) diffracted order of the array with 500 nm periodicity is measured at 538 nm. Coupling between the diffracted orders of the lattice and single-particle LSPRs results in well-known Fano lineshape asymmetry in the extinction spectra. The maximum extinction of the nickel nanoparticle arrays is larger than that of randomly distributed particles because of collective SLRs. The differences in shape and magnitude of the extinction signal are also reflected by the MCD spectra shown in Figs. 3(c). Since the MCD signal corresponds to the subtraction of two shifted extinction curves, its spectral features are more narrow and intense for the nickel
nanoparticle arrays. Again, the MCD null conditions are obtained at wavelengths that correspond to maxima in the optical extinction curves in Fig. 3(b), i.e. just above the diffracted order of the lattice. The MCD data of Fig. 3(c) clearly reveal that the perpendicular magnetization of nickel nanoparticles lifts the degeneracy of collective SLR modes that are excited by left- and right hand circularly polarized light.

Fig. 3. (a) Extinction spectrum (black curve, left scale) and MCD signal (red curve, right scale) of randomly distributed nickel nanoparticles. (b) Extinction spectra of nickel nanoparticle arrays with \( p = 400 \text{ nm} \) (black curve), \( p = 450 \text{ nm} \) (red curve), and \( p = 500 \text{ nm} \) (green curve). (c) MCD signal of the nickel nanoparticle arrays (same color coding as in (b)).

4. Modeling

To reproduce the optical extinction and MCD spectra of single nickel nanoparticles and periodic nanoparticle arrays, we first calculate the polarizability of the nickel nanoparticles using a model based on the modified long wavelength approximation (MLWA). This method
has been used in the past to successfully model optical [24–27] and magneto-optical
[15,28,29] properties of similar nano-sized cylinders. Because the particles are rotationally
symmetric and circularly polarized light can be decomposed into two linear orthogonal
components with a 90-degree phase difference, it is possible to describe the response of
nickel nanoparticles using a beam of linearly polarized light. The polarizability ($\alpha$) of a
cylindrical metallic nanoparticle with dielectric constant $\varepsilon_p$ embedded in a dielectric medium
with dielectric constant $\varepsilon_d$ can be approximated by extending the results of Meier and Wokaun
[30] to a spheroidal particle [31,32]

$$\alpha = \frac{V}{4\pi} \frac{\varepsilon_p - \varepsilon_d}{\varepsilon_d + L_{\text{eff}}(\varepsilon_p - \varepsilon_d)},$$

where $V$ is the volume of the particle and $L_{\text{eff}}$ is given by

$$L_{\text{eff}} = L - \frac{k^2}{3} \left( \frac{3V}{4\pi} \right) - \frac{k^3 V}{6\pi},$$

with $k$ being the wave vector of light ($k = 2\pi n/\lambda$). The first term in Eq. (2) is the geometrical
factor that accounts for the shape of the particle. For a spheroid, $L$ can be calculated
analytically [32]. For an oblate spheroid along its rotational axis (z-axis) it is given by

$$L_z = \frac{1}{e^2} (1 - \sqrt{1 - e^2}/e) \arcsin e,$$

where $e$ is the eccentricity of the spheroid. It is defined as $e^2 = (a^2 - c^2)/a^2$, with $a$ and $c$ being
the length of the major and minor semiaxes, respectively. For radiation along the $x$- or $y$-axis,
we take advantage of the sum rule $L_x + L_y + L_z = 1$ [33] and the rotational symmetry of the
system (i.e. $L_x = L_y$) to obtain

$$L_x = \frac{1 - L_z}{2}.$$  

The second term in Eq. (2) describes the dynamic depolarization that arises from dephasing
between radiation fields that are emitted from different parts of the particle. The last term
corresponds to a radiative reaction correction caused by recoil forces on accelerating charges.

We treat the MCD effect in a perturbative manner following the approach by Gu and
Kornev [34] and we introduce a magnetization-dependent dielectric constant $\varepsilon_p = \varepsilon_m \pm mQ_{\text{voigt}}$, where $Q$ is the magneto-optical Voigt constant of nickel [21] and $m$ is the magnetization of
the nanoparticle. The surrounding media (glass substrate and index matching oil) do exhibit
magneto-optical activity in an applied magnetic field because of diamagnetism. However,
since the effect is negligible compared to the magneto-optical response of the nickel
nanoparticles, it can be discarded. The polarizability of a single nickel nanoparticle can
therefore be written as

$$\alpha(m) = \frac{V}{4\pi} \frac{\varepsilon_m \pm mQ\varepsilon_m - \varepsilon_d}{\varepsilon_d + L_{\text{eff}}(\varepsilon_m \pm mQ\varepsilon_m - \varepsilon_d)},$$

Because of symmetry, reversal of the perpendicular magnetization in a nickel nanoparticle
has the same effect as a change of helicity in the incident light. MCD spectra can therefore be
obtained by calculating the difference in optical extinction for nanoparticles with
magnetization pointing up ($+m$) and down ($-m$). Using this notion, we calculate the
extinction cross section ($\sigma_{\text{ext}}$) of a nickel nanoparticle using
Here, the first and second term describe the contributions of optical absorption and scattering to \( \sigma_{\text{ext}} \), respectively. The relative difference in the extinction cross section for left- and right hand circularly polarized light is given by

\[
\Delta \sigma = \frac{\sigma_{\text{ext}}(+m) - \sigma_{\text{ext}}(-m)}{\sigma_{\text{ext}}(0)},
\]

where \( \sigma_{\text{ext}}(0) \) indicates the extinction cross section for a non-magnetized nanoparticle (i.e. \( Q = 0 \)). The parameter \( \Delta \sigma \) is proportional to the MCD effect. To compare the calculated extinction cross section (Eq. (6)) and \( \Delta \sigma \) (Eq. (7)) with the experimental results of Fig. 3, we scale both parameters to the maximum value of optical extinction in the experiments. The results for randomly distributed nickel nanoparticles are shown in Fig. 4(a). We find that the analytical MLWA model reproduces the experimental data well. The model also explains the asymmetrical shape of the MCD spectrum: The intrinsic magneto-optical Voigt constant of nickel [21] (\( Q \) in Eq. (5)) is larger at near-IR wavelengths, causing stronger MCD responses above the LSPR at 700 nm.

The MCD signal of the nickel nanoparticles originates mainly from the difference in optical absorption of left- and right hand circularly polarized light near the plasmon resonance condition. This is illustrated by Fig. 4(b), which depicts the calculated contributions from optical absorption and scattering to the extinction cross section and MCD spectra. The MCD signal that we obtain in our experiments is thus approximated by \( \text{MCD} = (E_+ - E_-)/E = (A_+ - A_-)/A \).

![Fig. 4. (a) Calculated extinction (black dashed curve, left scale) and MCD signal (red dashed curve, right scale) of randomly distributed nickel nanoparticles. For comparison, the experimental MCD signal is also shown (red solid curve). (b) Calculated contributions from optical absorption (red curves) and scattering (blue curves) to the extinction (black dashed curve, left scale) and MCD signal (black dashed-dotted curve, right scale). (c) Calculated extinction of a linear chain of nickel nanoparticles with \( p = 400 \) nm (black dashed curve), 450 nm (red dashed curve), and 500 nm (green dashed curve). (d) Calculated MCD signal of a linear chain of the nickel nanoparticles (dashed curves, same color coding as in (b)), together with experimental data (solid curves).](image-url)
To calculate MCD spectra of periodic nickel nanoparticle arrays, we extend the model using the coupled dipole approximation (CDA). Because of symmetry, we justifiably limit ourselves to the simplest possible case; a linear chain of nanoparticles. The polarization $\mathbf{P}_i$ of each particle with polarizability $\alpha_i$ at location $\mathbf{r}_i$ is given by

$$\mathbf{P}_i = \alpha_i \mathbf{E}_{\text{loc},i}(\mathbf{r}_i),$$

where $\mathbf{E}_{\text{loc},i}$ is the local electric field. This field includes contributions from the incident electric field and radiation that is emitted by other particles in the array. Normally, solving of Eq. (8) requires a self-consistent numerical approach. In the case of an infinite 1D particle chain, however, an analytical expression for the effective polarizability $\alpha_{\text{eff}}$ can be obtained if uniform polarization of the array elements is assumed [35].

$$\alpha_{\text{eff}} = \frac{\alpha_{sp}}{1 - S_{sp}}.$$  

Here, $\alpha_{sp}$ is the polarizability of a single particle given by Eq. (5) and $S$ is the retarded dipole sum that accounts for electric field radiation by other particles in the array. For a 1D particle chain, $S$ can be calculated using the method presented in [35,36]. In our case, the dipole sum is calculated for a particle chain of 1000 particles. For a lattice periodicity of 400 – 500 nm, this number of particles corresponds to a chain length of 400 – 500 µm, which is representative of the size of the periodic arrays in the experiments.

For single nickel nanoparticles, we determined the MCD effect by calculating the polarizability of the particles for opposite magnetization directions. We use the same approach for periodic nanoparticle arrays and insert the effective polarizability $\alpha_{\text{eff}}$ as defined by Eq. (9) into Eqs. (6) and (7) to obtain the extinction cross section and $\Delta \sigma$. Figures 4(c) and 4(d) show the resulting spectra after scaling to the maximum optical extinction in the experiments. Apart from the omission of the SLR that is associated with the diagonal $<1,1>$ diffracted order (for $p = 500$ nm), which is a direct consequence of using a 1D particle chain to estimate the response of a 2D lattice, the calculated spectra reproduce the experimental data well. The close correspondence is explained by the dominance of far-field diffractive coupling between nickel nanoparticles along the x- and y-axis of the periodic array and the symmetry of the system. The simple model used here is applicable to cylindrical magnetic particles in a square array.

Fig. 5. (a) Calculated extinction cross section of nickel nanoparticle arrays with $p = 500$ nm and different particle sizes. (b) Calculated relative difference in the extinction cross section for left- and right hand circularly polarized light. The arrays and color coding are the same as in (a).
5. Discussion

The MCD signal of nickel nanoparticle arrays originates from a small spectral shift between the extinction spectra (dominated by optical absorption) of left- and right hand circularly polarized light. Consequently, sizeable MCD effects are obtained at wavelengths where the slope of the extinction curve is large. From this notion, design rules for optimized MCD responses can be gathered. In the experimental spectra of Fig. 3(c), the first MCD peak of about −0.3% appears just after the diffracted order of the lattice. Here, the extinction sharply increases because of the excitation of a SLR. The second MCD peak of about + 0.4% is measured when the extinction drops after reaching its maximum value. Tuning of the width of spectral features in the extinction curves by, for example, variation of the nickel nanoparticle size would alter the MCD signal. To verify this effect, we calculated the extinction cross section and $\Delta \sigma$ for a fixed lattice periodicity of $p = 500$ nm and particle diameters varying from 60 nm to 180 nm. The results are summarized in Fig. 5. Obviously, the $\Delta \sigma$ signal (and thus MCD) of nickel nanoparticle arrays is highly tunable. Very sharp and intense spectral features are obtained for 60 – 100 nm particles near the diffracted order of the lattice (see inset). These narrow linewidth responses could be used for phase-sensitive detection in label-free biosensors, as recently illustrated by Kerr effect measurements on randomly distributed nickel nanoparticles [37]. Other routes that could be used to enhance the sensitivity of MCD-based sensors involve the use of magnetic materials with larger magneto-optical coupling strength $Q$ (enhancement of the MCD signal) or the integration of noble metal elements in magneto-plasmonic nanostructures (reduction of the plasmon resonance linewidth).

6. Conclusions

We have shown that the degeneracy of circular plasmon modes is lifted in cylindrical nickel nanoparticles. As a result, the extinction curves for left- and right hand circularly polarized light are shifted and a large MCD signal is measured near the single-particle LSPR condition. Coupling of the helicity-dependent LSPR modes to the diffracted orders of a nanoparticle lattice produces two narrow-linewidth SLR modes and an enhanced MCD signal. The MCD spectrum can be tuned via variation of the nickel nanoparticle size or the periodicity of the lattice. A simple numerical model based on the polarizability of spheroidal magnetic nanoparticles and coupling between dipoles in a 1D chain reproduces the main features of the experimental MCD spectra.

Acknowledgments

This work was supported by the Academy of Finland through the BENMAP project (No. 263510).