Experimental observation of the scattering of light by planar metallic nanoparticles

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Abstract. It is known that the efficiency of scattering of light by a particle is related to its size, geometry and optical constants of the material, since the theory of scattering by small particles developed by Mie in the beginning of the 20th century. However, the Mie scattering theory is valid just for a few special cases like a homogeneous sphere embedded on a medium of homogenous refraction index. More recently, some theoretical simulations on planar nanoparticles have shown that the optical resonances are dependent on the shape of the particle. Simultaneously, local field enhancements take place on the particles when excited by an incident wave.

A comprehensive knowledge of the optical behaviour of planar particles is of great importance for the development of optical devices at the nanoscale, as well as for the fast developing field of photonic crystals and the emerging field of diffractive optics.

In this paper, we present optical investigations on arrays of triangular nanoparticles with sizes comparable with the wavelength. Particles were produced using the method developed by Fischer and the optical measurements have been made using a scanning confocal microscope and a total-internal-reflection system.

The scattering of light by particles of some materials when excited by an evanescent field show a strong dependence on their relative orientation in the direction of incidence. The measured scattering patterns change with the polarization of light.
1. Introduction

The scattering of light by particles with sizes smaller than or comparable with the wavelength has been under intensive research. Control of the interaction of light with these particles and its associated effects are essential in the development of new optical devices at the micro- and nanoscale. Multiple scattering occurring in arrays of small particles or other regular nanostructures has become increasingly important in photonics, for example, in photonic crystals [1, 2].

Mie developed the first theoretical framework explaining the scattering and extinction of light by small spherical particles embedded in homogeneous environments [3]–[5]. The theory of Mie, however, does not take into account the multiple scattering that occurs in arrays of nanoparticles. It is restricted to isolated particles of a few geometrical shapes. More recently, Martin and Dereux [6] and others have tried to use a different approach based on the dyadic Green’s function to investigate the near-field of planar nanoparticles on structured substrates. Kottmann et al [7] have also shown that triangular silver nanoparticles smaller than the wavelength of light strongly enhance the field located at their corners, and that they have several resonances in their scattering cross-section unlike circular particles. The effect of size, shape and environment on the extinction efficiency has been discussed by Kelly et al [8].

The invention of the scanning near-field optical microscope (SNOM) [9] has allowed the investigation of the near-field of a sample and the provision of a higher optical resolution over the diffraction limit. It is still not clear, however, how this high-frequency spatial information is encoded in the optical signal [10]. Far-field techniques like the scanning confocal microscope, although limited to a maximum optical resolution of $\sim \lambda/2$, are much easier to interpret. Our microscope (alphaSNOM, commercialized by WITec GmbH) combines these two techniques into a single system together with an atomic force microscope (AFM). In this paper, we restrict our measurements to the confocal microscope for a more consistent interpretation.

It is well known that, in metallic materials, the quantized plasma oscillations (plasmons) are responsible for their optical behaviour. The plasmon confinement on nanoparticles generates resonances and local field enhancements [6]. One way to excite these plasmons and observe the resulting optical behaviour of the particles is to use a total-internal-reflection...
(TIR) excitation. The TIR has the advantage that the incident field does not superpose with the scattered field in the detection direction, unlike the cases of the reflection mode and transmission mode measurements. To understand how the shape and size of the particles influence their optical behaviour, we have produced arrays of nanoparticles of different sizes on glass substrates. We have investigated the interaction of a coherent light beam with the metallic nanostructures, using different excitation methods.

2. Sample preparation

The samples were prepared based on the colloid lithography method developed by Fischer and Zingsheim [11], usually called Fischer projection pattern (hereafter referred to as FPP). A monolayer of colloidal monodisperse polystyrene (PS) spheres was adsorbed on to a hydrophilic surface forming colloidal crystals. These crystals were later coated by evaporating a thin metallic film. By removing the layer of spheres after the evaporation, an array of triangular-shaped particles remains on the surface of the substrate. The linear size of the edge of the triangular particles depends mainly on the diameter of the spheres. Usually, the mean size of the triangular particles is smaller than the radius of the spheres (∼80% of the radius), but it depends also on the angle of evaporation relative to the substrate.

The literature contains several methods to produce self-assembled colloidal crystals, such as spin coating, dip coating, convective methods [12] and slow sedimentation with solvent evaporation [11, 13]. We have tested some of these methods but the best results have been obtained by using a slow sedimentation with solvent evaporation.

A drop of colloidal solution was put on the substrate (square cover slides of 24 × 24 mm² or 20 × 20 mm² and of a thickness of 170 µm) lying horizontally. With a micropipette, a small volume of a solution containing PS spheres was added on the glass surface and spread to the boundary of the substrate. A stationary state is reached in <5 min. The samples were kept in a plastic box of a few hundred cm³ to avoid air currents and maintain a better temperature equilibrium. The drying process takes between 2 h and up to 12 h, depending on the type of solvent and amount of liquid used, at a temperature of 20 °C. To obtain a good hydrophilic surface, the cover slides were modified by a plasma discharge for 10 s in a small glass container, at a pressure of ∼0.5 × 10⁻² mbar. This process increases the wettability of the surface.

We have used different solvents for the self-assembly process: methanol, ethanol and Milli-Q water. For spheres <0.5 µm, methanol was preferred, as the solution has a better homogeneity before the evaporation. However, water evaporates slowly and larger crystals can be formed.

Mainly three types of defects occur: partial multilayers, dislocations and vacancies. The dislocations are the most common defects and are extremely difficult to avoid. They occur after several small crystals are simultaneously formed and grow together as the evaporation proceeds. The dislocations start at some point of the crystal and increase their width along one of the crystal directions. The density and size of these defects depend on the evaporation rate and the dispersion of the diameter of the spheres. To form large crystals, it is therefore necessary to stabilize the solvent evaporation rate with a good temperature stability (±1 °C) and a narrow size distribution of the spheres. It is hence interesting to compare the optical behaviour of the defects with the behaviour of the more common triangular particles.

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2.1. PS spheres as lithographic masks

We have produced colloidal crystals using PS spheres commercialized by Polysciences Inc. The spheres were suspended in deionized water with a mass concentration of $\sim 2.5\%$ (kg m$^{-3}$). The nominal diameters of 3.176, 1.053, 0.528 and 0.338 $\mu$m, have been used to produce colloidal crystals.

Using the diameter of the spheres $D$, the area $A$ of the substrate surface to be coated and the concentration of spheres in the solution, it is possible to calculate the number of spheres necessary to cover the substrate with a monolayer. We have observed that these colloidal crystals form hexagonal close-packed arrays. Thus, to cover a substrate of area $A$ (m$^2$), using the hcp structure (for 2D) of a perfect crystal $N = 2A/\sqrt{3}D^2$, spheres of diameter $D$ (m) are necessary.

To obtain the concentration of solids in the solution $w$ ($\%$ kg m$^{-3}$ or $\%$ g ml$^{-1}$) and the specific mass of spheres $\rho$ (kg m$^{-3}$ or g ml$^{-1}$) it is necessary to use a liquid volume of $V = AD\rho\pi/(3\sqrt{3}w)$ (m$^3$). The amount of liquid used for the sedimentation method is a very important parameter in the quest to minimize the formation of multilayers and to form compact polycrystalline regions.

2.2. Film evaporation and lift-off of spheres

The samples with the adsorbed colloidal crystals were kept in a bell jar vacuum chamber for thermal evaporation. The vacuum pumping system, composed of a rotary pump and a turbo-molecular pump, reaches a base pressure of $\sim 10^{-6}$ mbar after 3 h. The pressure in the chamber is measured by a vacuum gauge unit, Leybold Combitron CM 330, using both Pirani and Penning sensors.

The evaporation system consists of one holder supporting an evaporation boat holding the material to be evaporated and another holder for the sample $\sim 22$ cm above the boat. Near the sample holder, there is a quartz oscillator crystal for monitoring the film thickness. The thickness of the evaporated films was measured by a Kronos QM 331 unit. An evaporation rate of 5–20 Å s$^{-1}$ was used.

After the evaporation, the samples were submitted to sonication in ethanol for a few seconds to remove the PS spheres. The spheres of smaller diameter ($< 356$ nm) were harder to remove from the surface. In those cases, a bath of toluene dissolves most of the PS and allows a faster and cleaner lift-off.

3. Topographical characterization

Topographical characterization of the samples has been made by two different methods: scanning electron microscopy (SEM) and AFM in tapping mode. The SEM was used to verify the size and organization of the colloidal crystals. An AFM (Digital Instruments Nanoscope IIIA AFM) working in tapping mode was used to characterize the topography of the FPPs. The AFM images have shown that the corners of the triangles are relatively sharp (radius of tenths of nm), but have a smaller film thickness compared with the middle. This is due to the process of evaporation. As the spheres act as masks, some material is adsorbed on to their edges. This reduces the evaporation flux through the interstices. Therefore the particles tend to form hills. This effect increases as the size of the PS spheres decreases and the evaporation
time increases. Figure 1 shows an SEM image of a colloidal crystal (left) and an AFM image of FPP.

### 4. Optical near- and far-field characterization

The optical properties, namely the reflection and transmission of monochromatic light by the FPPs reported above, have been studied using a microscope that combines the confocal and SNOM scanning techniques in the same system, although only confocal imaging has been used for the results presented here. One of the advantages of this system is the possibility of investigating the same sample in far-field using the confocal microscope and then investigate the near-field using the SNOM.

The scanning system is based on a scan table with capacitive sensors. These sensors allow lateral positioning to correct the typical drift of the piezos and to provide high linearity. From the fabricant specifications, a lateral resolution <1 nm can be obtained, which is sufficient for confocal and SNOM measurements.

Two lasers have been used as excitation sources for the confocal microscope: a He–Ne laser (\(\lambda = 632.8\) nm) with a mean power of 4 mW and an Ar-ion laser delivering six different wavelengths (\(\lambda = 457, 477, 488, 496, 502\) and 515 nm) with controlled output power up to 200 mW. The wavelength was selected by a holographic grating. Two mono-mode glass fibres couple the laser beams into the optical system of the microscope.

We use two sensors in the light intensity detection system: an SPCM-AQR APD coupled with a multi-mode fibre with a core diameter of 10 \(\mu\)m, used mostly for confocal detection in reflection mode, and a PMT (H6240) coupled with a fibre of 100 \(\mu\)m core diameter, used for the detection of the transmitted light.

The optical system is built around two objectives: an Olympus MPlan 100 \(\times\) NA = 0.90 used for confocal microscopy (excitation and reflected light detection) and a Nikon CFI 60 \(\times\) NA = 0.80 in the inverted microscope used for transmission measurements. The reflected light from the sample is spatially filtered by a pinhole before entering the fibre. This pinhole can be moved in two directions (X and Y) perpendicular to the reflected beam to achieve maximum signal intensity.

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Each image has been acquired with $512 \times 512$ pixel and a scan time of $1 \text{s line}^{-1}$. In the confocal microscope, we recorded only the intensity in the forward scan direction. The total scan time per image was more than 20 min, as some delay is recommended after each change of movement direction.

4.1. Confocal microscopy: reflection and transmission modes

Using the Rayleigh criterion, the maximum optical resolution that can be attained from an optical system is $d \geq 0.61 \lambda / (n \sin \theta)$, where $n \sin \theta$ is the numerical aperture (NA) of the objective. Thus, using a wavelength $\lambda = 457 \text{nm}$, a maximum resolution of $d_t \approx 350 \text{nm}$ for transmission and $d_r \approx 310 \text{nm}$ for reflection can be expected (see figure 2).

The FPP samples investigated have triangular particles with sizes of $\sim 400 \text{nm}$ (using PS spheres of diameter $1.053 \mu\text{m}$) and $\sim 1.2 \mu\text{m}$ (using spheres of diameter $3.176 \mu\text{m}$). Smaller FPPs have particles smaller than the optical resolution of the confocal microscope and, therefore, cannot be resolved. Samples prepared with PS spheres of $3.176 \mu\text{m}$ were preferred, since it is possible to identify the geometry of the particles and orientation of the array in the confocal microscope.

To evaluate better our confocal microscope images, we also measured some SNOM data. The SNOM images have a better resolution compared with the confocal images in transmission mode. We found that, for the rather large particle size used in this paper, no extra information on the scattering behaviour near the particles was detected. The SNOM was operated in illumination mode, using the tip as a local excitation source, in contrast with the PSTM, where the tip acts as a local probe of excitation induced by an evanescent field [14]. We would also mention that the tip of the SNOM cantilever was in contact with the sample. The measured data include the effect of an evanescent field interaction. A comparative study using different microscopes for particles of the same size and shape could help to explain the kind of influence the tip exerts on the optical behaviour of the particles.

Kottman et al [7] and Maas et al [15, 16] have shown that particles of triangular shape produce local field enhancements. These enhancements occur near the corners and some edges of the particles very close to them, showing a fast decay with the distance.
Figure 3. Set-up for confocal scanning microscope scattering experiment by TIR. The direction of the electrical field could be selected using a polarizer. For the p-polarization, the $E_\parallel$ vector was at an angle of $\sim 17^\circ$ with the surface of the sample.

However, the results of the calculations and measurements were obtained for particles of sizes $< 100$ nm. The particles we have produced have a mean size from hundreds of nm up to a few $\mu$m.

4.2. Experimental set-up for scattered light measurements by TIR

The geometry of the FPPs, where the film thickness is much smaller than the lateral dimensions of the particles, requires another type of excitation condition where the incident beam was parallel to the sample surface. This can be approximated by using a TIR excitation scheme (figure 3). In this case, the polarization state of the incident beam can be decomposed into the s- and p-polarized eigenstates.

The scattered light coming from the sample when excited by an evanescent field induced by TIR could be detected using the same objective used for confocal measurements in the reflection mode. Sequential scans of confocal measurements in reflection mode for vertical incidence and scattered light by TIR in the same sample region have been recorded to compare the optical behaviour by two different excitations.

A polarizer was placed before the prism to select the s-polarization (TE) and p-polarization (TM) states. The light source was a He–Ne laser coupled with a mono-mode glass fibre (NA = 0.12). At the end of the fibre, a collimator and a lens ($f = 150$ mm) were installed to focus the laser beam through the prism on to the sample surface to increase the power density.

When the sample was slightly misaligned with the scanning plane (an error of $1 \mu$m over a $50 \mu$m scan distance is sufficient), some fading occurred in the confocal measurements in the reflection mode. The confocal images obtained by TIR were less sensitive to this error. The wavelength used for TIR was 633 nm.
5. Dependency of scattering on the direction of incidence

The FPPs have a hexagonal configuration and most of the particles are approximately triangular. Two extreme cases with a rotation of 30° in-between can be distinguished due to symmetry: (a) all the six triangular particles of each hexagonal array have an edge perpendicular to the incident beam: three at the leading edge and three at the rear edge; and (b) all triangular particles have an edge parallel to the incident beam. Every other orientation is between these two extreme cases.

The optical contrast of aluminium particles on glass in the visible range of the spectrum is very high. Aluminium films have a complex refraction index of \( \tilde{n} = n + ik \approx 1.15 + i6.39 \) for \( \lambda = 632.8 \text{ nm} \) [17, 18], although these values also depend on the method of fabrication. Several FPPs of aluminium were produced using PS spheres of diameter 1053 and 3176 nm. The thickness of the evaporated films was \(~20\) nm. For aluminium films, this thickness was larger than the skin depth and almost no light was transmitted. On a bare glass surface, only 4% of the light was reflected and, therefore, a good optical contrast was obtained.

In the samples with FPPs generated with spheres of a nominal diameter of 1053 nm, we detected the dependence of scattering on the orientation of the crystals relative to the incidence beam. As each sample contains many crystals of several hundreds of \( \mu \text{m}^2 \), any orientation can be obtained, similar to those obtained by rotating a single crystal. However, the size of the particles (\(~400\) nm) did not allow a clear identification of the scattering location. The samples with FPPs using PS spheres of \(~3\) \( \mu \text{m} \) allow us to locate the scattering patterns.

The scattering patterns of aluminium nanoparticles produced using PS spheres of diameter \(~3\) \( \mu \text{m} \) have shown high sensitivity to the orientation relative to the incident beam. We have measured the intensity of scattered light for two different orientations, as described above, for s- and p-polarization. For the p-polarization, the electrical field was not completely perpendicular to the sample, but had a small component parallel to the sample surface in the incidence direction (figure 3). The incident beam inside the prism was at an angle of 17° with the sample surface. The ratio of the parallel and perpendicular components of the electrical field relative to the sample surface was \(~0.3\).

In each case, a confocal image was obtained using a vertical incidence source (\( \lambda = 457 \text{ nm} \)) to locate the particles. Using the same scanning region and the same focal plane of the objective, the TIR image was recorded thereafter. The scanned region was almost the same, with only a small lateral drift.

Figures 4 and 5 show the confocal images in reflection mode and their respective TIR patterns for s- and p-polarization observed for an aluminium FPP. Figure 4 has been obtained using the first orientation of the crystal relative to the incident beam, as described above, whereas the images shown in figure 5 correspond to the second case.

For each TIR image, the vectors \( S = E \times B \) and \( E \) are displayed on the top right of the figure.

The TIR image (figure 4) with p-polarization exhibits strong scattering centres, located at the edges of the triangular particles which have their rear edge perpendicular to \( S \). At the same time, the other three particles show two lobes in the direction of incidence but of much weaker intensity. Particles of approximately square shape have their scattering centres concentrated at opposite corners located along the \( Y \)-direction. Some of them also have a lobe near the left corner. Particles with four edges and a more elongated shape show a
Figure 4. Confocal scanning microscope images for vertical and TIR illumination of an aluminium FPP. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

Figure 5. Confocal scanning microscope images for vertical and TIR illumination of an aluminium FPP, where the crystal orientation was rotated 30° relative to figure 4. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

more complex scattering pattern. Several lobes are located near the two edges located on the left.

In the s-polarization case, the scattering patterns are different: triangular particles with their rear edge perpendicular to $S$ have a weak scattering lobe (in some cases two) located near this edge. The other three have a strong scattering centre located at the leading edge and a small lobe of weaker intensity on the left of the first. Particles with four edges have the scattering concentrated at the opposite corners in the direction of the electrical field.

It must be noted that in the p-polarization case, the vector $E$ is not perpendicular to the plane of the sample. The component of the electrical field parallel to the sample has a smaller intensity compared with the vertical one, but may be responsible for the double-scattering lobes located on three of the triangular particles in each hexagon. A similar effect could be responsible for the lobe located at the left corner of some particles of square shape. It is not clear why this scattering pattern does not occur for the other three particles.
When the crystal in figure 4 is rotated by 30°, every triangular particle has an edge parallel to the plane of incidence and almost parallel to \( S \). This is the other extreme case that can be obtained for hexagonal symmetry.

Again, we have observed different scattering patterns for this crystal orientation. For both s- and p-polarization, the scattering centres of higher intensity on the triangular particles are located at their corners oriented in the \( Y \)-direction.

For p-polarization, only a few triangular particles show smaller lobes. Their additional lobe is located on the left of the particle. Particles with approximately square shape have a high-intensity lobe located at their left edge.

In the s-polarization case, the triangular particles have two more lobes of weaker intensity located on the upper left and right or on the lower left and right. In contrast, particles of approximately square shape have a lobe of high intensity located at their right edge and a region of weaker intensity located on the left.

Some of the remarkable scattering patterns observed can be explained by the dynamics of the electron plasma and its local confinement, driven by the incident wave. In figure 4, for p-polarized light, the two lobes in the direction of incidence located on three of the particles of each array may be due to a dipolar interaction. There is a component of the electrical field in this direction. The other three particles do not show a similar intensity. For the s-polarization, the scattering behaviour is more symmetrical. It should be mentioned that the particles with the right edge perpendicular to \( S \) are stronger scatterers when compared with particles with the left edge in this orientation.

The lobes located at the opposite corners in the approximately square shape particles are easier to explain in the image of s-polarized light. In this case, the electrons are driven by a wave parallel to the surface of the particle and there is a charge concentration near the opposite corners. For p-polarized light, the scattering patterns are more complex.

According to Jackson [19], when the size of the scatter is comparable with or larger than the wavelength, a multipole analysis of the scattering is required. Kelly et al [8] have also reported that multipolar scattering is connected to the size of a particle. However, the results shown in this paper were obtained only for isolated spheres in vacuum. A numerical simulation for particles of this shape and size, under the illumination conditions reported here, could give a more complete explanation for the scattering patterns observed.

6. Dependence of scattering on the material

We tried to find out how the optical constants of the planar particles influence the scattering patterns. To study the influence of the dielectric constant, FPPs of similar geometry using materials with different refraction indices \( \tilde{n} \) have been produced.

FPPs of gold (a noble metal) and SiO (an insulator) have been produced following a similar procedure as for aluminium. The film thickness was 20 nm for both cases. Gold has a complex refraction index \( \tilde{n} = n + i\kappa \approx 0.20 + i3.32 \) [17, 18] and SiO has \( \tilde{n} = n + i\kappa \approx 1.95 + i0.01 \) [20] at \( \lambda = 632.8 \) nm. It should be noted that the values presented here may not correspond exactly to the films we have produced. For SiO, for example, when the evaporation takes place in a vacuum >\( 10^{-6} \) mb, many oxygen atoms are adsorbed and change the stoichiometry of the SiO films. Therefore the SiO films are specified as SiO\(_x\), with \( x \) lying between 1 and 2.
Figure 6. Confocal scanning microscope images for vertical and TIR illumination of a gold FPP. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

Figure 7. Confocal scanning microscope images for vertical and TIR illumination of a gold FPP, with a crystal rotation of 30°. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

Confocal and TIR images of the gold FPP are shown in figure 6. Similar scattering patterns as in the case of aluminium (figure 4) were found. Here, the lobes with higher intensity seem to be more spherical compared with figure 4. For p-polarized light, however, there are remarkable differences: every triangular particle has a lobe located at its left edge, with one or more smaller lobes on the left. The other important feature is the relatively high intensity at the corners of the particles of approximately square shape when compared with figure 4.

For the second orientation of the gold FPP, scattering patterns similar to the aluminium case have been found, at least for s-polarized light. The main differences are lobes located at the particles with four edges and an elongated shape (s- and p-polarization) and the fading towards the upper left observed in the triangular particles of lobes. The last feature could be due to the small inclination between the crystalline orientation (vector $\langle 10 \rangle$) and the plane of incidence (see figure 7).

SiO FPPs with small film thickness have a low optical contrast as can be observed in figures 8 and 9. The same is true for the TIR measurements, although the structures and
the local scattering can be identified. Samples with thicker films (for e.g. 40 nm) were also investigated. The optical contrast of the reflection images was better, but the scattering features in TIR measurements were similar.

For both the crystal orientations, SiO FPPs show a weak dependence on the polarization direction. But in the first case (figure 8), one can see that the same triangular particles (with the rear edge perpendicular to the plane of incidence) have lobes of higher intensity for both polarizations. Evidently, this is in contrast to the previous TIR images for gold and, especially, for aluminium.

Although no other insulator material has been tested, it seems that the observed dependence of the scattering on polarization is strongly associated with the complex refraction index present in metals. In insulators, the extinction index $\kappa$, or the corresponding absorption coefficient is very small and can be neglected. The scattering depends on the difference in the dielectric constant between the particle and the environment.

One experiment that can be done is to produce SiO$_2$ FPPs on glass and to investigate the scattering by TIR. In this case, in a direction perpendicular to the sample, there is no dielectric

**Figure 8.** Confocal scanning microscope images for vertical and TIR illumination of a SiO FPP. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

**Figure 9.** Confocal scanning microscope images for vertical and TIR illumination of a SiO FPP, with a crystal rotation of 30°. Left: confocal image obtained in reflection mode (vertical incidence), $\lambda = 457$ nm; middle: confocal image with p-polarized TIR illumination, $\lambda = 633$ nm; right: confocal image with s-polarized TIR illumination, $\lambda = 633$ nm.

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contrast between the substrate and the FPP particle, but in a direction parallel to the FPP there are SiO$_2$ particles surrounded by air. If the topography profile is high enough, the scattered light can be measured.

7. Conclusion

We have shown that the scattered light by TIR of FPPs of planar metallic nanoparticles of gold and especially aluminium present a strong dependence on their orientations relative to the incidence direction of light and the polarization. The geometry of the particles also determines the location of the scattering lobes.

We have observed significant scattering differences among the FPPs of different materials. Our investigations were done on particles with the same type of interface: glass substrate and air. Therefore, we cannot infer the influence of the dielectric contrast between the particles and the environment. To investigate how the dielectric constant influences the scattering patterns, more materials have to be investigated and a wide range of wavelengths have to be studied. Results of Kelly *et al* [8] show that the substrate changes the location of the resonances towards longer wavelengths.

Kottmann *et al* [7] obtained numerical results for isolated silver particles of much smaller size. Nonetheless, it is clear how geometry affects the scattering cross-section and the local electrical field distribution. We have not observed the field distributions predicted by Kottmann *et al*, but it should be stressed that the size of the particles and the illumination conditions are, except for the s-polarization case, different. For s-polarization, we have reported high scattering intensities at the corner or at the edge of the triangular particles, depending on their relative orientation.

With the SNOM microscope, the smaller features and the finer structure of the lobes could be investigated. However, the SNOM measures an evanescent field, whereas the confocal microscope measures the intensity of a propagating wave. The SNOM may yield extra information when used in collection mode, under TIR illumination conditions. In this case, the results obtained by SNOM and confocal microscopy could be compared. The difference between the use of SNOM in illumination mode as reported by Maas *et al* [15, 16], and our set-up is the polarization state. In our case, non-polarized light was used at vertical incidence. The pyramidal tip configuration and the optical set-up do not allow us, at the moment, to use the SNOM tip as a polarized light source.

Another interesting aspect is the dependence of the scattering centre’s location on the dielectric constant of the material of the particles. The numerical results presented by Maas *et al* [16] indicate an increasing pattern complexity when going from dielectric to noble metal. We have also reported that the gold and aluminium FPP particles show more complex patterns compared with SiO and the scattering is very sensitive to the direction of incidence. However, the particles we have investigated have a different size.

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