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Evidence of multipolar excitations in surface enhanced Raman scattering

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Visible and near-infrared extinction spectra of regular arrays of gold nanorods display several bands depending on polarization of the electromagnetic incident field. When setting the incident field perpendicular to the nanorod axis (*transverse polarization*), a single band is observed, which is assigned to a dipolar plasmon mode. The several bands obtained under an incident polarization along the nanorod axis (*longitudinal polarization*) are attributed to multipolar plasmon resonances. Raman scattering experiments carried out under transverse and longitudinal laser excitations on methylene blue adsorbed on these arrays show that multipolar plasmon resonances can give rise to strongly enhanced spectra with an amplification per molecule similar to that obtained for dipolar plasmon excitation.

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I. INTRODUCTION

Surface enhanced Raman scattering (SERS), observed for the first time in 1974 on a silver roughened electrode,[1] has become an ultrasensitive spectroscopy since, under optimal conditions, it enables one to detect a single molecule adsorbed onto a single particle.[2,3] It is now well established that the origin of SERS comes from electromagnetic[4] (em) and to a lesser extent from electronic (chemical), contributions.[5–7] Additional enhancement can occur if the molecule under consideration undergoes an electronic transition, the absorption band of which coincides with the laser excitation wavelength (resonant SERS: SERRS).

Extensive theoretical studies have been developed to describe the experimentally observed Raman amplification of up to 10^{14}.[2,3] Former theories involving em enhancement deal with a molecule assumed to be a polarizable point dipole close to a single noble metal sphere[8] or spheroid.[9] The excitation of localized surface plasmon by the incident laser beam leads to an enormous amplification of the incident and Raman scattered electromagnetic fields. These calculations yield a Raman gain of the order of 10^5–10^6 and are thus unable to account for the detection of a single molecule. More recent calculations consider a molecule trapped into a microcavity formed by two or several electromagnetically interacting particles generating huge electromagnetic fields.[10–16] Such calculations lead to the concept of active sites (*hot spots*) for which the Raman amplification can reach ten orders of magnitude. The strong inhomogeneity of the electromagnetic field within such cavities brings about multipoles of high order. For instance, the calculated Raman enhancement for the pyridine molecule located between two silver spheres was found to reach ten orders of magnitude provided a multipole expansion of the electromagnetic field up to the 48th order is taken into account.[16]

However, these calculations are unable to account for the observation of strong SERS signals when excitation takes place *off plasmon resonance*,[2,17,18] even if it has been recently shown that the hot spot resonance profile is redshifted with respect to the main plasmon resonance.[15]

To our knowledge, even if some calculations have shown the importance of multipolar resonances in SERS mechanisms,[19,20] up to now no experiments have been undertaken to estimate the contribution of high order multipolar plasmon excitations to the Raman gain. Since it has been recently shown that elongated nanorods exhibit spectrally well separated multipolar plasmon modes,[21–24] we show in this paper the ability of multipolar plasmon resonances of such nanorods to generate SERS spectra.

II. EXPERIMENTAL TECHNIQUES

Regular arrays of gold nanorods are prepared by electronic beam lithography (EBL) according to the procedure previously described.[25] Briefly, the first step is to deposit by spin-coating (60 s, at 5000 rpm) onto indium tin oxide (ITO)–doped glass (10×10 mm) a 100-nm-thick layer of polymethyl-methacrylate (PMMA). After exposure of the substrate to EBL equipment writing [JEOL 6400 scanning electronic microscope (SEM) supplied with a RAITH quantum pattern generator] and chemical developing (PMMA developer, Allresist Comp.), a 45-nm gold film is thermally evaporated at a 1-nm/s rate. The remaining PMMA is *lifted off* with acetone leaving the particles on ITO at sites defined by EBL exposure. The length, width, and height of the gold nanorods are then checked by SEM and atomic force microscopy (AFM). The height h and the cross section axis length l of the nanorods are kept approximately constant to 45 nm and 110 nm ± 10 nm, respectively, while the long axis length L was set to the following values: 210 nm (array A), 260 nm, 270 nm, 300 nm, 340 nm, 390 nm, 420 nm, 450 nm, 480 nm, 490 nm, 550 nm (array B), and 650 nm. The spacing between the ends of two consecutive nanorods is fixed to 110 nm ± 10 nm, and is 300 nm ± 20 nm in the perpendicular direction.

Extinction spectra are recorded using a LOT-ORIEL model MS260i spectrometer (focal length 260 mm,

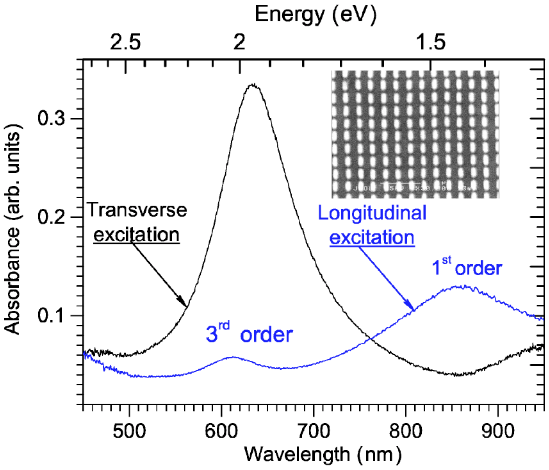

FIG. 1. Extinction spectra of gold nanorod array A ($L=210$ nm, $l=110$ nm $h=45$ nm) recorded in air under transverse and longitudinal polarizations. Spectrum recorded under longitudinal polarization exhibits two bands assigned, respectively, to the first- ($\lambda_{\mathrm{long}}^{(1)} \simeq 870$ nm) and third-order ($\lambda_{\mathrm{long}}^{(3)} \simeq 610$ nm) plasmon resonances, while the transverse one shows a single band assigned to the dipolar mode ($\lambda_{\mathrm{trans}}^{(1)} \simeq 630$ nm). Inset: Scanning electronic microscopy (SEM) image of the array.

150 grooves/mm grating giving a spectral resolution of approximately 5 nm) equipped with a 1024 diode array thermo electrically cooled at 0 °C. This spectrometer is coupled by fiber optics to a Leitz microscope supplied with ×20 magnification lens of a 0.30 numerical aperture (NA). The light source is a halogen lamp illuminating the sample with a collimated beam. The investigated zone is a circle of approximately 40 μm diameter smaller than the array size (squares of 100×100 μm).

Raman spectra are obtained at three excitation wavelengths (514, 647, and 676 nm) using a triple stage DILOR XY microspectrometer equipped with a charge coupled device (JOBIN-YVON, 1024×256 pixel CCD, thermoelectrically cooled at 160 K). Excitation wavelengths are provided by a 2018 model Spectra-Physics Ar-Kr laser. The molecular probe considered in this paper is methylene blue (MB) dye, which yields a tiny fluorescence[26] (0.02 ± 0.005 quantum yield) and displays a very strong absorption band at approximately 675 nm, which can give rise to resonance surface-enhanced Raman scattering provided laser excitation takes place within its absorption band. Raman spectra of MB are excited by focusing the laser beam (Gaussian shape of 2 mm full width at half maximum) on the array, and recorded using backscattering geometry and a ×100 magnification microscope objective of NA 0.80. Arrays are first dipped into $10^{-4}M$ aqueous solution of methylene blue, then thoroughly washed to remove molecules not directly adsorbed onto the array and dried with nitrogen gas. It is then assumed that only a single layer of MB dye remains adsorbed onto the array according to the observation of no further change in the Raman spectrum upon further rinsing.

To allow the comparison of the SERS enhancement for various plasmon excitations, it is useful to define the Raman enhancement G per molecule as the ratio of the intensity in presence and in absence of the array[27]

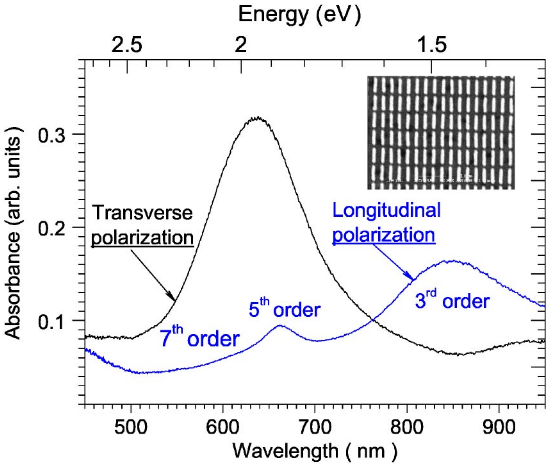

FIG. 2. Extinction spectra of gold nanorod array B ($L=550$ nm, $l=110$ nm $h=45$ nm) recorded in air under transverse and longitudinal polarizations. Spectrum recorded under longitudinal polarization exhibits two bands assigned, respectively, to the third- ($\lambda_{\mathrm{long}}^{(3)} \simeq 850$ nm) and fifth-order ($\lambda_{\mathrm{long}}^{(5)} \simeq 660$ nm) plasmon resonances, while the transverse one shows a single band assigned to the dipolar mode ($\lambda_{\mathrm{trans}}^{(1)} \simeq 635$ nm). Inset: Scanning electronic microscopy (SEM) image of the array.

$$G = \frac{N_2}{N_1}\frac{I_{\mathrm{SERS}}}{I_{\mathrm{ref}}}. \qquad (1)$$

In this equation, N_1 is the number of molecules covering the array within the laser spot area, and N_2 the number of molecules excited in the reference sample. Consequently, I_{SERS}/N_1 is the SERS integrated intensity of some typical band reduced to a single adsorbed molecule, and I_{ref}/N_2 the corresponding spontaneous Raman intensity of the same band. Considering only a single layer, the number of molecules under laser spot of 5 μm^2 area is $N_1 \approx 6.2 \times 10^6$ (0.8 nm^2 area). The scattering volume for the reference samples was evaluated to be 5000 μm^3,[27] such that the number N_2 of molecules in the reference sample is 3×10^{10} for the $10^{-2}M$ solution of MB in water. The enhancement per molecule was thus evaluated using the following relation:

$$G = 4.8 \times 10^3 \frac{I_{\mathrm{SERS}}}{I_{\mathrm{ref}}}. \qquad (2)$$

III. RESULTS AND DISCUSSION

A. Extinction spectra

Examples of extinction spectra of gold nanorod arrays (labeled hereunder as A and B, respectively) are given in Figs. 1 and 2. These spectra are recorded with the incident electric field polarized perpendicular (*transverse polarization*) and parallel (*longitudinal polarization*) to the nanorod axis. Under transverse polarization, the single band observed is assigned to the first order (dipolar) plasmon excitation. The fact that only the dipolar plasmon band appears in the visible range is consistent with the rather small nanorod cross section ($l \approx 110$ nm).[28] Under longitudinal polarization,

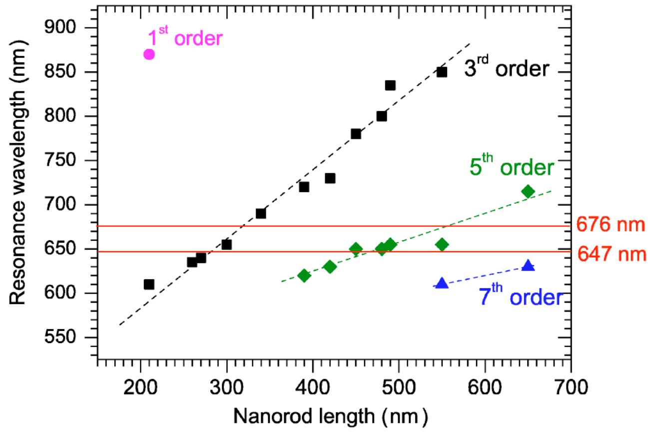

FIG. 3. (Color online) Plasmon resonance wavelengths of various multipolar orders as a function of nanorod length for spectra recorded in air. The nanorod height and short axis are 45 nm and 110 nm, respectively, while the long axis is varied from 210 nm to 650 nm. Lines drawn between experimental points represent the best linear fit. Horizontal lines represent the laser excitations at 647 nm and 676 nm.

several bands appear in both spectra which are attributed according to the literature[21,22] to the excitation of plasmon modes of odd multipolar order: $\lambda_{\text{long}}^{(1)} \simeq 870$ nm, $\lambda_{\text{long}}^{(3)} \simeq 610$ nm for array A, and $\lambda_{\text{long}}^{(3)} \simeq 850$ nm, $\lambda_{\text{long}}^{(5)} \simeq 660$ nm for array B. When increasing nanorod length, plasmon resonances of higher multipolar order appear in the visible range, while lower ones are shifted towards infrared with a progressive broadening reflecting retardation effects and radiation damping.[21–24,28] These results are summarized in Fig. 3, where longitudinal plasmon resonance wavelengths are plotted versus the nanorod length: As previously observed,[21–23] the resonance wavelengths increase faster with nanorod length for lower multipolar orders, thus making easy the band assignments.

B. Raman spectra

We now turn our attention to Raman spectra of MB adsorbed onto these arrays in order to investigate the possibility of obtaining spectra arising from dipolar or multipolar plasmon resonance using transverse or longitudinal laser excitations respectively. These experiments can be performed by merely a 90° rotation of the sample while keeping the same laser polarization and excitation wavelength since the third and fifth plasmon resonances of arrays A and B are located in a spectral region (610, 660 nm, respectively) neighboring the wavelength of dipolar resonance under transverse polarization (630, 635 nm, respectively).

Methylene blue dye displays its major Raman bands in the 1200-1700 cm^{-1} spectral region. Figure 4 shows that the intensity of the Raman bands in this spectral range is almost the same for transverse and longitudinal excitations at 647 nm. Spectra obtained on array B using 676 nm laser excitation (Fig. 5) shows again that transverse and longitudinal excitations give rise to similar Raman intensity. The Raman intensity appears slightly lower for excitation at 676 nm with respect to that obtained at 647 nm excitation because of the weaker laser power available at 676 nm (see caption of Fig. 5). When taking spectra outside the arrays, no signal can be detected, which demonstrates that the spectra do arise from SERS. When using laser excitation at 514 nm, only tiny intensity spectra can be obtained according to the fact that excitation takes place outside any plasmon resonance. Enhancement factors per molecule calculated with Eq. (2) for the different arrays using 647 nm and 676 nm excitations are summarized in Table I. The relative uncertainty evaluated from standard deviation of several measurements is evaluated to 25%. Raman spectra obtained using 514 nm excitation are too weak to enable any valuable evaluation of the enhancement factor per molecule.

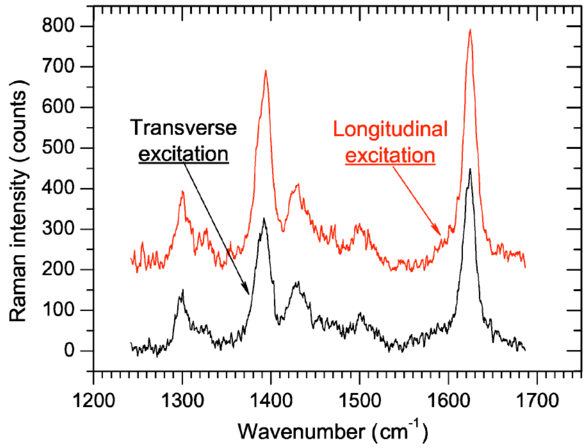

FIG. 4. Raman spectra of methylene blue excited under transverse and longitudinal excitations at 647 nm on array A. Laser power 25 mW at the sample, slit widths 300 μm, counting time 30 s. Spectra are vertically shifted for clarity.

As a general result (Table I), the order of magnitude of the enhancement factor per molecule is about 10^5, which is similar to that previously obtained on gold nanoparticles regular arrays.[27,29] On account of the estimated relative uncertainty (25%) on the enhancement factor, we can merely conclude to

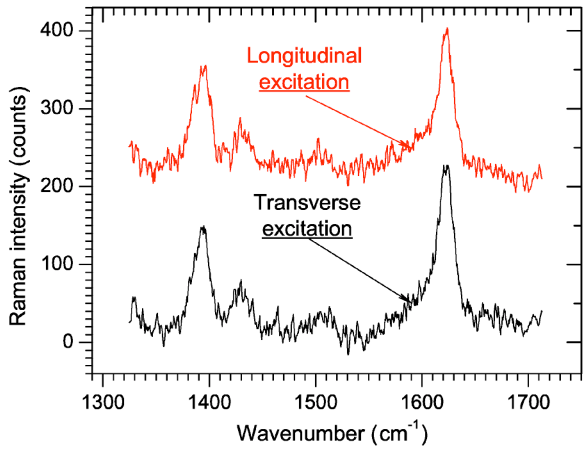

FIG. 5. Raman spectra of methylene blue excited under transverse and longitudinal excitations at 676 nm on array B. Laser power 16 mW at the sample, slit widths 300 μm, counting time 30 s. Spectra are vertically shifted for clarity.

TABLE I. Enhancement factor per molecule obtained on the different arrays transverse or longitudinal laser excitations. The enhancement relative uncertainty evaluated from the standard deviation of several measurements is about 25%.

	Nanorod length (nm)					
Excitation wavelength (nm)	210	260	270	300	340	390
Transverse 647	1.5×10^5	1.1×10^5	2.2×10^5	2.3×10^5	1.5×10^5	1.4×10^5
Longitudinal 647	1.3×10^5	7×10^4	2.0×10^5	1.9×10^5	1.3×10^5	2.1×10^5
Transverse 676	1.3×10^5	1.2×10^5	1.8×10^5	1.3×10^5	1.0×10^5	1.1×10^5
Longitudinal 676	8×10^4	8×10^4	1.4×10^5	1.4×10^5	8×10^4	1.0×10^5

	Nanorod length (nm)					
Excitation wavelength (nm)	420	450	480	490	550	650
Transverse 647	1.8×10^5	2.1×10^5	1.8×10^5	1.2×10^5	2.0×10^5	1.3×10^5
Longitudinal 647	2.7×10^5	2×10^5	2×10^5	1.7×10^5	1.4×10^5	2.0×10^5
Transverse 676	1.1×10^5	1.0×10^5	1.1×10^5	1.8×10^5	9×10^4	1.4×10^5
Longitudinal 676	1.1×10^5	1.2×10^5	1.3×10^5	1.1×10^5	9×10^4	1.6×10^5

a comparable enhancement for transverse and longitudinal polarizations.

We question now about the plasmon resonance at the origin of the Raman enhancement. When the laser polarization is set perpendicular to the nanorod axis, only a single band is observed in the extinction spectrum (Figs. 1 and 2). The band maximum is always located in the same spectral region at about 630–640 nm, according to the similar cross section for all the investigated nanorods. This resonance plasmon should be assigned to the dipolar excitation of the plasmon of a single nanorod on the ITO substrate and not to a *grating-induced mode* which may appear at metal dielectric interface when the single particle plasmon resonance over the dielectric index $(\lambda_{\mathrm{LSP}}/n)$ approaches the grating constant.[30] This result is consistent with obtaining a comparable enhancement factor on all the arrays, reflecting the fact that the Raman enhancement does proceed from the resonant excitation of individual dipolar plasmon resonance. Extinction spectra (Figs. 1 and 2) recorded under longitudinal polarization display several bands that have been identified on similar arrays as arising from the excitation of plasmon resonances of odd multipolar order.[21–24] When the polarization of the incident electromagnetic field is oriented along the nanorod axis, the dipolar coupling occurs perpendicular to it, that is, along a direction for which the interparticle spacing (grating constant) is kept constant to 300 nm ± 20 nm in all the arrays. For such grating constant, the near-field and far-field dipolar coupling are weak; besides, on account of its much shorter range, multipolar coupling is negligible with respect to dipolar one. This enables us to conclude that the Raman enhancement should arise from the excitation of multipolar plasmon of individual nanorods when the laser polarization is set along the nanorod axis. However, one may question about a possible Raman enhancement resulting in a coupling between the longitudinal excitation and the transverse one through either the particle multipolar or Raman scattered fields. Plasmon resonance of order greater than one does not radiate efficiently, so that the plasmon coupling from multipolar order should be very weak. The Raman scattered light is totally depolarized such that any coupling that could occur with plasmon modes would be similarly distributed over dipolar and multipolar modes. Lastly, the Raman intensity found, when the laser polarization is set at 45° of the nanorod axis, lies between the intensities obtained under 0° and 90° incident angles. This is consistent with the assumption that in this latter case, both longitudinal and transverse modes are excited. We may thus conclude that the enhancement of Raman spectra recorded under longitudinal laser excitation does proceed mainly from multipolar plasmon resonances of individual nanorods.

To support this conclusion, we have taken extinction and Raman spectra under oblique incidence angles. Changing the incident angle when the laser polarization is set within the incident plane (TM or *p* polarization) and along the nanorod axis leads to the excitation of even order multipoles that appear as additional bands in the extinction spectra.[23] Such examples of even multipolar plasmon resonances are given in Fig. 6 for an array of gold nanorods of 450 nm length. While for perpendicular incidence only modes of odd orders are excited, additional extinction bands appear for oblique incidence $(\theta=20°)$.[23] These bands are attributed to plasmon modes of even order. For further inclination $(\theta=40°)$, only even order modes are favorably excited.[23] This figure also shows that the 647 nm laser excitation lies within the fourth order plasmon resonance under 40° incidence angle and between the third and fifth order for 0° incidence. According to this figure, we should expect that the intensity of the Raman spectrum recorded under 40° incidence angle should be significantly greater than that at 0°. This is indeed what it is observed in Fig. 7 where the Raman spectrum under excitation at 40° incidence angle is about twice more intense than that at 0°.

IV. CONCLUSION

In this article, we investigated extinction spectra of various length gold nanorod arrays and Raman spectra of methylene blue dye adsorbed as a single layer on these arrays. When the incident electric field is polarized perpendicular to the nanorod axis, a single plasmon band is observed which is

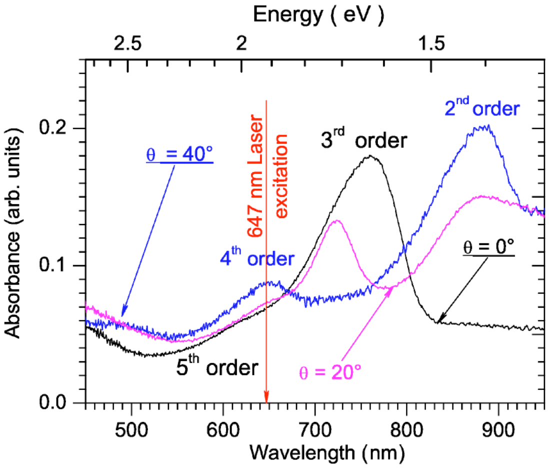

FIG. 6. (Color online) Extinction spectra recorded on a regular array of gold nanorod with axis length $L=450$ nm, transverse axis $l=110$ nm, and height $h=45$ nm. Under normal incidence, only plasmon resonance of odd multipolar order appear. When inclinating the sample at 20° under TM or p polarization along the nanorod axis, even order plasmon bands appear. Under 40° incidence angle, only even order plasmon resonances remain visible due to an unfavorable excitation (Ref. 23).

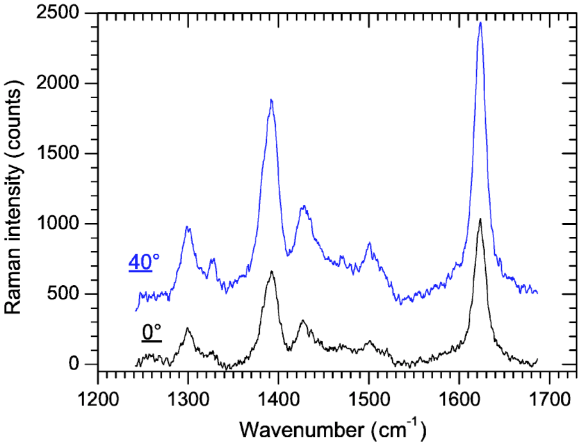

FIG. 7. Raman spectra of methylene blue dye adsorbed on the nanorod array of axis length $L=450$ nm, transverse axis $l=110$ nm and height $h=45$ nm recorded with 647 nm laser excitation at 0° and 40° incidence angles. Laser power 24 mW at the sample is focused with a ×50 magnification objective of NA 0.55, counting time 60 s, slit widths 300 μm. Spectrum at incidence angle of 40° is about twice more intense than that at 0° due laser excitation within the plasmon resonance of fourth order.

attributed to dipolar plasmon resonance. Under an electric field polarization along the nanorod axis, several bands are observed which intensity and wavelength depend on the nanorod length. These bands are assigned accordingly to the literature[21–24] to plasmon resonances of odd multipolar order. Raman spectra of methylene blue dye adsorbed as a single layer on these arrays are recorded under transverse and longitudinal polarizations leading to SERS spectra of comparable intensity. The extensive Raman measurements performed on nanorod arrays leads us to conclude that the Raman enhancement proceeds from dipolar or multipolar plasmon resonances depending on whether the laser excitation is polarized perpendicular or parallel to the nanorod axis. Besides, this Raman enhancement is found to be of the same order of magnitude for parallel and perpendicular polarizations. This latter observation enables us to suggest an explanation for the very strong Raman enhancement that can occur for aggregated particles even when the excitation takes place in the infrared, far from the first order plasmon resonance.[2,17,18] We propose that this very strong enhancement may result from the excitation of high multipolar plasmon resonances located in the infrared. These high multipolar resonances may be generated within microcavities that are formed by the aggregation process and result from the strong inhomogeneity of the electric field within the microcavities.[14]

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