Active gold nanoshells for surface enhanced Raman scattering

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1. Introduction

Surface enhanced Raman scattering (SERS) relies on the large localized electric fields near nanoscaled metal surfaces due to the optical excitation of their surface plasmons (SPs) [1]. In metal nanoparticle, the G factor can reach about $10^{11}-10^{12}$ when the molecules are trapped at the hotspots [2]. But, the hotspot region in whole nanosystem is very small and hence a low SERS efficiency. The metal nanoparticle incorporating gain media can be called as the active nanoparticle. The gain materials transfer energy to compensate the loss of SP and finally amplify the desired SP response. Recently, Li et al. [3] have introduced the gain media into the core of cubic gold nanoboxs and obtained an extremely high SERS G factor on $10^{16}-10^{17}$.

2. General Instructions

We investigate the electric field enhancements of the gold nanoshell with gain-doped dielectric core. The radii of the inner core and outer shell r_1 and r_2 are fixed at 40 and 50 nm, respectively. For simplicity, a complex refractive index for the inner core is set as n-ik. n is fixed at 1.43 for host medium of SiO₂ and gain coefficient is related to the amount of optical gain induced by external pumping [3]. The surrounding medium is assumed as air. As k = 0, a strong peak appears at about 639.5 nm, which is due to the dipole resonances in the gold nanoshell. When the k-value increases to a critical value at 0.4188, a super SP resonance happens in the active gold nanoshell at about 629.5 nm. The peak intensity of $Q_{\rm sca}$ reaches about 1.91×10^7 , which is about 8.16×10^6 times to that for k = 0. We also find that the Q_{abs} shows a great negative enhancement. The negative Q_{abs} -value means that the scattered radiation is amplified by the energy transferred from the gain media [4]. In the passive nanoshell, the larger enhancements of the electrical fields occur along the incident polarization and only locate within a few nanometers of the outer shell surface, which can be well interpreted by the dipole plasmon resonance. The maximal G-value is about 5500. In the active gold nanoshell, the distribution of the electric fields in the active gold nanoshell is similar to that observed in the passive gold nanoshell. An enormous enhancement of the electric fields is found in the active gold nanoshell, which is not realized in the previous report [4]. The maximal G-value can reach about 3.25×10^{17} .

We further investigate the influence of the shell thickness on the electric field enhancements in the active gold nanoshells. The radius of inner core is fixed at 40 nm. With increasing the thickness of the gold shell, the wavelength of the super-resonance in active gold nanoshell shows a blue shift. At the same time, the gain coefficient for super resonance decreases first from 0.745 at $r_2 - r_1 = 2$ nm to 0.3893 at $r_2 - r_1 = 8$ nm and then increases to 0.548 at $r_2 - r_1 = 15$ nm. Especially, it is found with increase in shell thickness that the maximal G-value obtained in the active gold nanoshells increases first from 7.15×10^{17} at $r_2 - r_1 = 2$ nm to 7.62×10^{19} at $r_2 - r_1 = 8$ nm and then decreases to 7.67×10^{15} at $r_2 - r_1 = 15$ nm. When the shell thickness is very thin, the coupling between the two modes is very strong and hence the high metallic losses. Under this condition, the larger gain threshold is due to the higher metallic losses. The increased thickness of gold shell will lead to the decrease of the critical value of gain coefficient. On the other hand, the increased thickness of the gold shell increases the gold component and hence the increased metallic losses. The competition between two mechanisms causes the minimum of the gain coefficient at $r_2 - r_1 = 8$ nm. It is further found with increase in the d-value that the G factor decreases from 7.62×10^{19} at d = 0 nm to 3.48×10^{16} at d = 50 nm. d indicates the distance away from the outer surface. The large hotspot region means that this optimized active gold nanoshell possesses high SERS efficiency.

3. Conclusions

The electric field enhancement properties of the active gold nanoshell have been investigated. As the gain coefficient increases to a critical value, a super resonance has been found in the active gold nanoshell, which induces the enormous enhancement of the electric fields. An extremely high SERS G factor on $10^{19}-10^{20}$ can be obtained in the optimized active gold nanoshell. The optimized active gold nanoshell has a high efficiency of SERS effect and may be well applied for the single molecule detection.

References

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Appendix

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