

## Article

# Effect of Rod-like Nanoparticles on the Dielectric Susceptibility of Nematic Nano-Composites: A Molecular Theory

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**Abstract:** The effect of rod-like nanoparticles on the high-frequency dielectric susceptibility of the nematic nano-composites has been investigated in the framework of a molecular theory. Analytical expressions for the components of the effective polarizability of a rod-like nanoparticle in the nematic host have been obtained and used in the calculations of the dielectric susceptibility of the composites as functions of the nanoparticle volume fraction. Numerical calculations of the susceptibility have been undertaken using the nematic liquid crystal 5CB as a host doped with either gold or silver particles for different values of the concentration of nanoparticles. It has been shown that the rod-like nanoparticles have a much stronger effect on the components of the dielectric susceptibility of the nano-composites including, in particular, the one with gold nanoparticles in the vicinity of the plasmon resonance. The main conclusion is that at sufficiently large concentration of nanoparticles, the anisotropy of the dielectric susceptibility of the nano-composites may even change the sign with an increasing concentration which may be important for various applications.

**Keywords:** liquid crystal; molecular-statistical theory; nano-composite



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## 1. Introduction

At present, there exists a significant interest in liquid crystal (LC) nano-composites in which the nematic host phase is doped with various types of nanoparticles (NPs) including dielectric, metal or semiconductor ones. It has been shown that doping of the nematic phase with such NPs can affect many important properties of the nematic materials including switching voltages and switching times of LC displays [1–5]. Doping of nematic LCs with ferroelectric NPs may also enhance dielectric and optical anisotropy and the electro-optical response [6,7] as well as the photorefractive properties [8]. Nematic nano-composites with para- and ferromagnetic NPs can be used to develop magnetically tunable structures, while doping of ferroelectric LCs with metal and silica nanoparticles may lead to the increase of the spontaneous polarization and dielectric permittivity [9–11]. On the other hand, the dielectric properties of NPs may be significantly modified in the LC medium. For example, it is possible to tune the plasmon resonance of gold NPs by changing the dielectric properties of the liquid crystal host phase [12–14].

In the previous paper [15], we have developed a molecular theory of the dielectric susceptibility of nano-composites doped with spherical nanoparticles. It has been shown that in the range of resonant frequencies, the nanoparticles significantly affect the dielectric susceptibility of the nematic host phase. In this paper, we have used the model of a hard long metal rod to derive explicit expressions for the effective polarizability of rod-like NPs in the nematic host. These expressions have then been used to evaluate the components of the high-frequency dielectric susceptibility of the nematic composites for small NP volume fractions. Finally, the high-frequency dielectric susceptibility of the nematic LC 5CB (4-cyano-4'-pentylbiphenyl) doped with gold and silver NPs has been calculated numerically

as function of frequency. It will be shown that long rod-like metal nanoparticles can make an even stronger effect on the dielectric susceptibility of the nano-composite, and, in particular, on its anisotropy which may even change the sign at some NP concentration.

The main objectives of this paper are as follows. (i) To develop a molecular theory of the high frequency dielectric susceptibility of the nematic nano-composite doped with rod-like nanoparticles (Section 2.1). (ii) To obtain explicit expressions for the effective polarizability of rod-like gold and silver nanoparticles in the nematic host and to calculate numerically the components of the dielectric susceptibility of the nematic nano-composites for a particular nematic host doped with gold and silver nanoparticles (Section 2.2).

## 2. Results

### 2.1. High-Frequency Dielectric Susceptibility of the Nematic Nano-Composite

The nano-composite can be expressed using the Clausius–Mossotti equation where the polarizabilities of a mesogenic molecule and of the NP are replaced by the effective renormalized polarizabilities that depend on intermolecular correlations between NPs and mesogenic molecules [15–17]:

$$\frac{(\hat{\varepsilon} - \hat{I})}{(\hat{\varepsilon} + 2\hat{I})} = \frac{4\pi}{3} \left( (1 - \phi)\hat{\beta}_m^{eff} + \phi\hat{\beta}_{NP}^{eff} \right), \quad (1)$$

where  $\hat{I}$  is a unit tensor and  $\phi$  is the volume fraction of the NPs. On the right-hand side of Equation (1), the first term is the sum of the effective nondimensional polarizability of the mesogenic molecule and that of the NP (i.e., the polarizabilities normalized by the corresponding molecular volume).

In this paper, we consider a nematic phase doped with a small volume fraction of NPs. Then, the effective molecular polarizability  $\hat{\beta}_m^{eff}$  is approximately expressed in terms of the dielectric susceptibility  $\hat{\varepsilon}_{LC}$  of the pure nematic phase:

$$\frac{(\hat{\varepsilon}_{LC} - \hat{I})}{(\hat{\varepsilon}_{LC} + 2\hat{I})} = \frac{4\pi}{3} \hat{\beta}_m^{eff}. \quad (2)$$

Thus, Equation (1) can be rewritten in the form:

$$\frac{(\hat{\varepsilon} - \hat{I})}{(\hat{\varepsilon} + 2\hat{I})} = \frac{(1 - \phi)(\hat{\varepsilon}_{LC} - \hat{I})}{(\hat{\varepsilon}_{LC} + 2\hat{I})} + \frac{4\pi}{3} \phi \hat{\beta}_{NP}^{eff}. \quad (3)$$

Equation (3) is, in fact, a linear equation in terms of  $\varepsilon$ , and hence, it can readily be solved to yield the following expression for the dielectric tensor of the composite:

$$\hat{\varepsilon} = \frac{\hat{\varepsilon}_{LC} + \frac{2\phi}{3} \left[ \hat{I} - \hat{\varepsilon}_{LC} + \frac{4\pi}{3} \hat{\beta}_{NP}^{eff} (\hat{\varepsilon}_{LC} + 2\hat{I}) \right]}{\hat{I} + \frac{\phi}{3} \left[ (\hat{\varepsilon}_{LC} - \hat{I}) - \frac{4\pi}{3} \phi \hat{\beta}_{NP}^{eff} (\hat{\varepsilon}_{LC} + 2\hat{I}) \right]}. \quad (4)$$

In the case of small concentration of NPs, Equation (4) can be expanded in powers of small NP volume fraction  $\phi$ , keeping the linear terms:

$$\hat{\varepsilon} \approx \hat{\varepsilon}_{LC} - \phi(\hat{\varepsilon}_{LC} - \hat{I}) + \frac{4\pi}{3} \phi \hat{\beta}_{NP}^{eff} (\hat{\varepsilon}_{LC} + 2\hat{I}), \quad (5)$$

where the susceptibility tensor of the nematic host phase is expressed in terms of the nematic director  $\mathbf{n}$ :

$$\hat{\varepsilon}_{LC,\alpha\beta} = \varepsilon_{\perp} \delta_{\alpha\beta} + (\varepsilon_{\parallel} - \varepsilon_{\perp}) n_{\alpha} n_{\beta}. \quad (6)$$

where  $\alpha, \beta$  are the Cartesian coordinates  $x, y, z$ . The first term in Equation (6) describes a decrease of the dielectric susceptibility due to a decrease of the volume fraction of mesogenic molecules replaced by NPs. In contrast, the second term in (6) describes the

contribution from NPs. The results of our previous paper [15] indicate that this contribution is predominant.

## 2.2. Effective Polarizability of a Rod-like Nanoparticle in the Nematic Phase

In this section, we use the model of a strongly anisotropic NP which is assumed to be an ellipsoid of revolution filled with the isotropic metal of the susceptibility  $\varepsilon_{NP}$  which is embedded in the nematic liquid crystal medium with the susceptibility  $\hat{\varepsilon}_{LC}$  given by Equation (6). Then, the electric field  $\mathbf{E}^{NP}$  inside the ellipsoid is homogeneous and can be expressed as [18]:

$$\hat{\varepsilon}_{LC}(\hat{I} - \hat{q})\mathbf{E}^{NP} + \hat{q}\mathbf{D}^{NP} = \hat{\varepsilon}_{LC}\mathbf{E}_m, \quad (7)$$

where the induction  $\mathbf{D}^{NP} = \varepsilon_{NP}\mathbf{E}^{NP}$  and  $q_{\alpha\beta} = q_{\perp}\delta_{\alpha\beta} + (q_{\parallel} - q_{\perp})a_{\alpha}a_{\beta}$ . Here,  $q_{\parallel}, q_{\perp}$  are the depolarization coefficients of the ellipsoid of revolution defined, for example, in [18]; the unit vector  $\mathbf{a}$  is in the direction of the axis of the ellipsoid.

Thus, the electric field inside the ellipsoid can be expressed as:

$$\mathbf{E}^{NP} = \frac{\hat{\varepsilon}_{LC}\mathbf{E}_m}{(\hat{\varepsilon}_{LC} - \hat{\varepsilon}_{LC} \cdot \hat{q} + \varepsilon_{NP}\hat{q})}. \quad (8)$$

The dipole moment of the ellipsoid in the dielectric medium can be expressed as:

$$\mathbf{P} = V(\mathbf{P}^{NP} - \mathbf{P}_m), \quad (9)$$

where

$$\mathbf{P}^{NP} = \frac{1}{4\pi}(\varepsilon_{NP} - 1)\mathbf{E}^{NP}, \quad \mathbf{P}_m = \frac{1}{4\pi}(\hat{\varepsilon}_{LC} - \hat{I})\mathbf{E}_m, \quad (10)$$

where  $V$  is the NP volume.

On the other hand

$$\mathbf{P} = \hat{\alpha}^* \cdot \mathbf{E}_m, \quad (11)$$

where  $\hat{\alpha}^*$  is the effective polarizability of the ellipsoid in the nematic LC dielectric medium.

Substituting Equations (8) and (10) into Equation (9) and then into Equation (11), one obtains the following general expression for the effective polarizability of the ellipsoidal NP in the tensor form:

$$\hat{\alpha}^* = \frac{V}{4\pi} \frac{\hat{\varepsilon}_{LC}(\varepsilon_{NP}\hat{I} - \hat{\varepsilon}_{LC})}{(\hat{\varepsilon}_{LC} - \hat{\varepsilon}_{LC} \cdot \hat{q} + \varepsilon_{NP}\hat{q})}. \quad (12)$$

One notes that Equation (12) expresses the effective polarizability of the model NP in any coordinate system and for any orientation of the ellipsoid. Assuming for simplicity that the long axis of the ellipsoidal particle is perfectly ordered along the director of the nematic phase (i.e.,  $\mathbf{a} = \mathbf{n}$ ), one obtains the following expressions for the transverse and the longitudinal components of the effective polarizability:

$$\alpha_{\perp}^* = \frac{V}{4\pi} \frac{\varepsilon_{\perp}(\varepsilon_{NP} - \varepsilon_{\perp})}{(\varepsilon_{\perp} - \varepsilon_{\perp} \cdot q_{\perp} + \varepsilon_{NP}q_{\perp})}. \quad (13)$$

$$\alpha_{\parallel}^* = \frac{V}{4\pi} \frac{\varepsilon_{\parallel}(\varepsilon_{NP} - \varepsilon_{\parallel})}{(\varepsilon_{\parallel} - \varepsilon_{\parallel} \cdot q_{\parallel} + \varepsilon_{NP}q_{\parallel})}. \quad (14)$$

Finally, for sufficiently long rod-like NPs,  $q_{\perp} = 1/2$  and  $q_{\parallel} = 0$ , and hence, one obtains the following expressions for the components of the effective polarizability of rod-like NPs:

$$\alpha_{\perp}^* = \frac{V}{2\pi} \frac{\varepsilon_{\perp}(\varepsilon_{NP} - \varepsilon_{\perp})}{(\varepsilon_{\perp} + \varepsilon_{NP})}. \quad (15)$$

$$\alpha_{\parallel}^* = \frac{V}{4\pi} (\varepsilon_{NP} - \varepsilon_{\parallel}). \quad (16)$$

Substituting Equations (15) and (16) into Equation (5), one obtains the approximate expressions for the transverse and the longitudinal components of the dielectric susceptibility of the nematic nano-composite:

$$\varepsilon_{\perp}^{NC} \approx \varepsilon_{\perp} - \phi(\varepsilon_{\perp} - 1) + \frac{2}{3}\phi \frac{\varepsilon_{\perp}(\varepsilon_{\perp} + 2)(\varepsilon_{NP} - \varepsilon_{\perp})}{(\varepsilon_{\perp} + \varepsilon_{NP})}, \quad (17)$$

$$\varepsilon_{\parallel}^{NC} \approx \varepsilon_{\parallel} - \phi(\varepsilon_{\parallel} - 1) + \frac{1}{3}\phi(\varepsilon_{\perp} + 2)(\varepsilon_{NP} - \varepsilon_{\perp}). \quad (18)$$

One notes that the dielectric susceptibility of the nano-composite depends on the components of the dielectric susceptibility of the host nematic phase and on the susceptibility  $\varepsilon_{NP}$  of the medium inside the ellipsoidal NP.

In this paper, we use the extended Drude model for a rod-like metal NPs with a plasmon resonance. The susceptibility of gold is described by using a combination of the Drude model and two critical point terms [19].

$$\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + i\gamma\omega} + \sum_{CP=1}^2 G_{CP}(\omega), \quad (19)$$

where  $\omega_p$  is the plasma frequency,  $\gamma$  is the collision frequency and

$$G_{CP}(\omega) = A_{CP}\Omega_{CP} \left( \frac{e^{i\varphi_{CP}}}{\Omega_{CP} - \omega - i\Gamma_{CP}} + \frac{e^{-i\varphi_{CP}}}{\Omega_{CP} + \omega + i\Gamma_{CP}} \right), \quad (20)$$

where  $G_{CP}(\omega)$  are the contributions from the interband transitions (gaps),  $A_{CP}$  is the amplitude,  $\Omega_{CP}$  is the frequency of the gap,  $\varphi_{CP}$  is the phase, and  $\Gamma_{CP}$  is the broadening [20]. The coefficients  $A_{CP}$ ,  $\Omega_{CP}$ ,  $\varphi_{CP}$ , and  $\Gamma_{CP}$  are given in [19].

The permittivity of silver is described by using a combination of the Drude model with one Lorentz term

$$\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + i\gamma\omega} + \frac{(\varepsilon_s - \varepsilon_{\infty})\omega_r^2}{\omega^2 + i\Gamma_0\omega}, \quad (21)$$

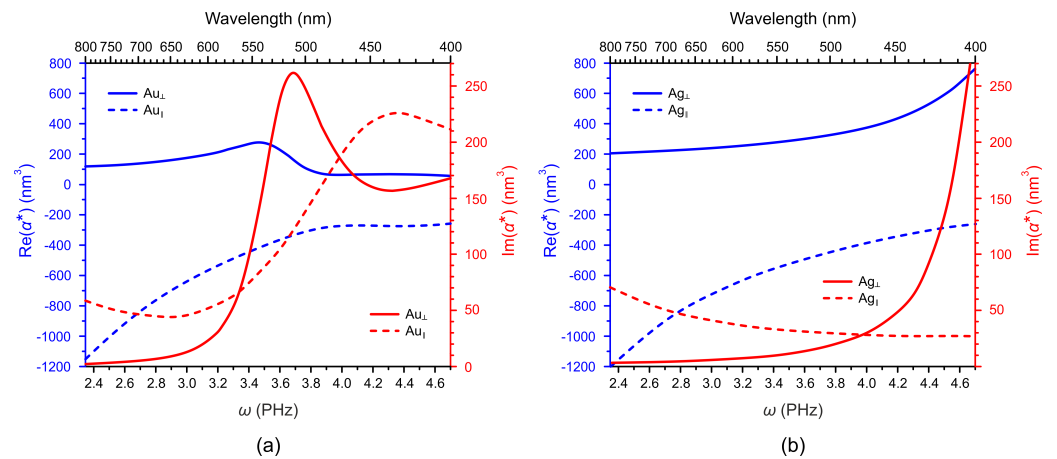
where  $\omega_r$  is oscillator resonant frequency,  $\Gamma_0$  is the damping factor, and

$$\varepsilon_s = \varepsilon_{\infty} + \frac{\omega_p^2}{\omega_r^2}. \quad (22)$$

The coefficients  $\varepsilon_{\infty}$ ,  $\omega_p$ ,  $\gamma$ ,  $\omega_r$ , and  $\Gamma_0$  are calculated from experimental data [21] by means of the Horiba-Jobin-Yvon DeltaPsi 2 software.

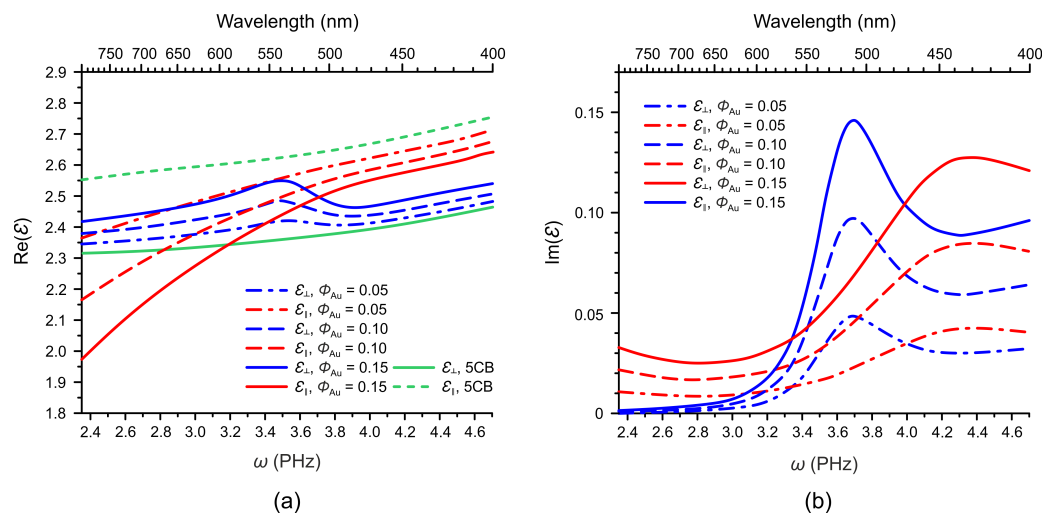
Both approximations showed good agreement with the experimental results from [21].

The components of the effective polarizability of the gold NPs (AuNPs) and silver NPs (AgNPs) in 5CB are presented in Figure 1. The calculations have been made using the corresponding data for gold and silver and for the nematic 5CB [22] at room temperature. One notes that both longitudinal and transverse susceptibilities of 5CB have been measured in the selected frequency range [22]. At the same time, the resonance frequency of both spherical and rod-like AuNP is within the selected frequency range while that of the AgNP is outside the range for both types of NPs.

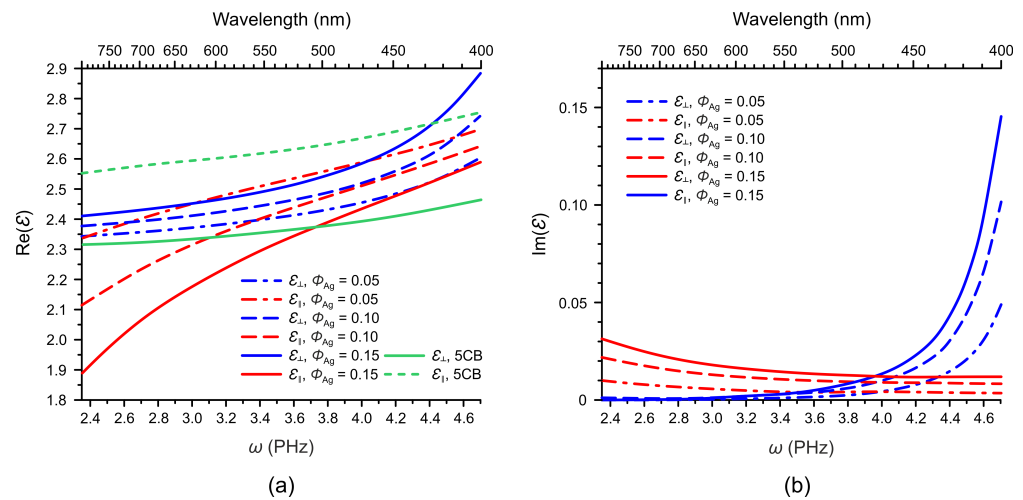


**Figure 1.** Real and imaginary parts of the transverse and longitudinal components of the effective high-frequency polarizability of gold (a) and silver (b) rod-like NPs embedded into the 5CB nematic host phase. Rod-like NP is an ellipsoid of revolution with the principal diameters 35 nm and 5 nm.

Longitudinal and transverse components of the dielectric susceptibility of the 5CB nematic nano-composites doped with gold and silver nanoparticles are presented in Figures 2 and 3 as functions of frequency for different values of the volume fraction  $\phi$  of the NPs. Similar to the case of spherical nanoparticles [15], one can readily see the splitting of the plasmon resonance in nano-composites with rod-like gold NPs and the shift of the resonance determined by the increase of the volume fraction  $\phi$ . At the same time, one notes that long rod-like gold NPs have a much stronger effect on the dielectric susceptibility of the nematic nano-composite compared with the spherical gold NPs [15]. In particular, the anisotropy of the susceptibility appears to be very sensitive to the concentration of rod-like NPs. One notes that the significant difference between the pattern for AuNP and AgNP composites is mainly determined by the fact that the resonance frequency of the AgNP is outside the range where the frequency dependence of the dielectric susceptibility of the nematic host 5CB has been measured, being located at higher frequencies. Thus, we cannot extend the frequency range of the diagram but one expects that the corresponding pattern around the resonance frequency of the AgNP composite will be qualitatively similar to that of the AuNP one.

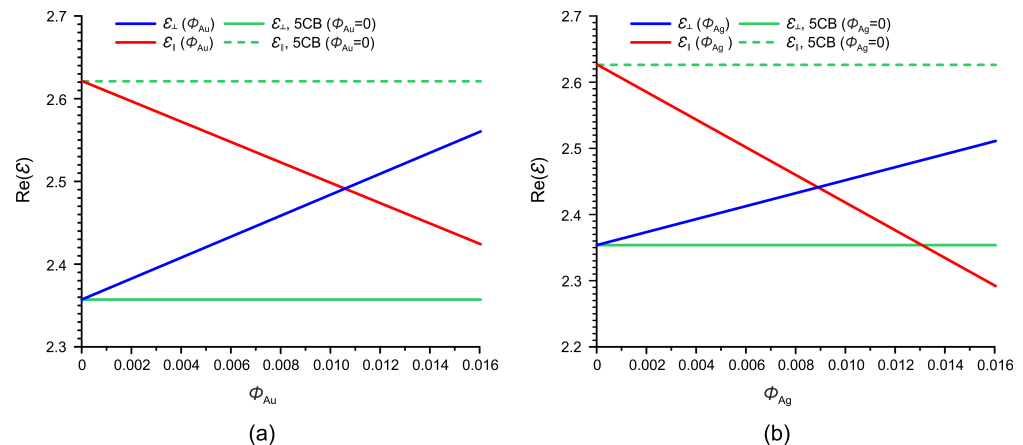


**Figure 2.** Real (a) and imaginary (b) parts of the transverse and longitudinal components of the high-frequency dielectric susceptibility of the 5CB nematic nano-composites for different values of the volume fractions of AuNPs.



**Figure 3.** Real (a) and imaginary (b) parts of the transverse and longitudinal components of the high-frequency dielectric susceptibility of the 5CB nematic nano-composites for different values of the volume fractions of AgNPs.

Finally, in Figure 4, we present the dependence of the real parts of the longitudinal and transverse components of the dielectric susceptibility of the 5CB nematic nano-composite on the volume fraction of gold and silver NPs. It is interesting to note that the dielectric anisotropy of the nano-composite is decreasing with increasing NP volume fractions. Moreover, the anisotropy changes the sign for both composites with gold and silver NPs at some relatively small value of NP concentration and becomes negative at larger NP concentration. This property can also be very interesting from an application point of view.



**Figure 4.** Real parts of the transverse and longitudinal components of the high-frequency dielectric susceptibility of the 5CB nematic nano-composites as functions of the volume fraction of gold (a) and silver (b) nanoparticles for the cyclic frequency of 3.5 PHz.

### 3. Conclusions

Analytical expressions for the effective anisotropic polarizability of a rod-like NP in the nematic LC host phase have been obtained. The effective NP polarizability has then been used to evaluate the high-frequency dielectric susceptibility of the corresponding nematic nano-composites with long rod-like gold and silver NPs in the case of a small NP volume fraction. In contrast to the case of spherical NPs, considered in our previous paper [15], where a simple semi-phenomenological expression for the effective isotropic polarizability in the isotropic medium has been used, we have explicitly taken into account the dielectric anisotropy of the nematic LC medium surrounding the rod-like NP.

In this paper, we have also employed the extended Drude model for rod-like metal NPs with a plasmon resonance and compared it with a simple Drude model used in [15].

In particular, the permittivity of gold has been described by using a combination of the Drude model and two critical point terms [19], while the permittivity of silver is described by using a combination of the Drude model and one Lorentz term. The parameters of the models have been calculated from experimental data [21] and both approximations have demonstrated good agreement with the experimental results.

The components of the dielectric susceptibility of the nematic nano-composites based on 5CB LC doped with rod-like gold and silver NPs have been calculated numerically as functions of frequency for different values of NP volume fraction. Similar to the case of spherical NPs [15], the splitting and the shift of the plasmon resonance have been observed determined by the change of the volume fraction of rod-like NPs. At the same time, it has been shown that at least in the resonance frequency range, long rod-like gold NPs have a much stronger effect on the components of the dielectric susceptibility of the nematic nano-composite compared with the spherical gold NPs. In particular, the dielectric anisotropy of the nano-composite is decreasing with increasing volume fraction of both gold and silver NPs, and it may even change the sign at some relatively small value of NP concentration. This may be important for optical applications of nematic nano-composites. In general, the rod-like metal NPs are expected to be very effective in controlling the optical and dielectric properties on nematic nano-composites.

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