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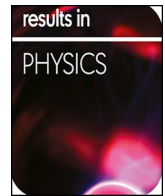
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# Topological magnetoelectric effect as probed by nanoshell plasmonic modes

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## ABSTRACT

Axion electrodynamics is applied to study the response of a plasmonic nanoshell with a core made of topological insulator (TI) materials. The electric polarizability of such a system is calculated in the long wavelength limit via the introduction of two scalar potentials satisfying the various appropriate boundary conditions. Our focus is on the topological magneto-electric effect (TME) as manifested in the coupled plasmonic resonances of the nanoshell. It is found that for a TI with broken time-reversal symmetry, such TME will lead to observable red-shifts in the coupled plasmonic modes, with more significant manifestation of such shifts for the bonding modes of a metallic nanoshell. It is speculated that such universal red-shift could be a manifestation of the fundamental dual symmetry as generalized for axion electrodynamics.

## Introduction

The concept of the hypothetical particle – axion – was first introduced in the late 1970's to account for the “strong CP problem” [1–3], which was later found also relevant for the dark matter problem [4–6]. Its connection to electrodynamics was first formulated by Sikivie in his proposition of using electromagnetic coupling for the detection of the elusive particle [7]. This was followed up by Wilczek in his application of axion electrodynamics to account for the possible fractional charge on dyons and its possible relevance to the quantized Hall effect [8]. Moreover, it was not until the late 2000's that axion electrodynamics (AED) was ultimately realized in the then newly-discovered topological insulators.

Topological insulators (TI) are quantum states of (2D/3D) matter with an insulating interior but conducting edge/surface states, with these boundary conducting states being protected topologically by time-reversal symmetry [9,10]. It was first recognized in 2008 that the origin of such topological boundary states, namely, the quantum spin Hall effect, can be accounted for in a phenomenological way with the application of AED, leading to the so-called topological magneto-electric (TME) response of the system [11]. Such electromagnetic/optical response of the TI in turn leads to intriguing optical effects such as Faraday and Kerr rotations of incident polarized beams at THz frequencies, which were only recently confirmed in a series of ingenious experiments by various groups [12–14].

Since AED is formulated as a modified Maxwell electromagnetic theory with the inclusion in the Lagrangian of the pseudo scalar product term  $(\vec{E} \cdot \vec{B})$  between the fields, it will have implication to all phenomena in classical optics other than the Faraday and Kerr polarization rotations, such as the modified Fresnel reflection from TI studied in previous literature [15]. In particular, scattering from localized TI targets will have a potential to reveal unique TME effects. Indeed, such scattering process from TI target has been studied intensively in recent times for different geometries of the target and in different external environments [16–19]. For example, it was observed that TME-induced strong backward scattering can take place from certain configuration of targets made of TI cylinders [18]. Moreover, in most of the previous studies of scattering off a localized TI target, only bare target particles have been considered, leaving unexplored with the possibility of using composite TI particles for the revelation of unique scattering signatures from TME effects.

It is the purpose of our present work to study electromagnetic interaction with a plasmonic nanoshell containing a TI core to explore possible unique TME signatures which may provide alternative approach to the study of such universal TME effects. As is well-known in the literature, since its fabrication in 1998, plasmonic nanoshell has been recognized as a highly-versatile system based on its coupled-plasmon modes arising from the outer and inner interfaces of the shell. In time, various applications ranging from opto-thermal cancer therapy [20] to powerful optical sensing technology [21] have been developed.

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Here our goal is to investigate the shifts of the coupled resonances from these nanoshells with the core replaced by a TI material, limiting ourselves to subwavelength particles. The results obtained can hence be revealed via Rayleigh scattering [18,19] from such composite nanoshells.

### Theory

The modified electromagnetic field equations as applied to TI was first formulated by Qi et al [11] and can be expressed in the following form (in Gaussian units):

$$\begin{aligned} \vec{\nabla} \cdot \vec{D} &= 4\pi\rho - 4\pi\kappa \vec{\nabla} \theta \cdot \vec{B} \\ \vec{\nabla} \cdot \vec{B} &= 0 \\ \vec{\nabla} \times \vec{E} + \frac{1}{c} \frac{\partial \vec{B}}{\partial t} &= 0 \\ \vec{\nabla} \times \vec{H} - \frac{1}{c} \frac{\partial \vec{D}}{\partial t} &= \frac{4\pi}{c} \vec{J} + 4\pi\kappa \left[ \frac{1}{c} \frac{\partial \vec{B}}{\partial t} + \vec{\nabla} \theta \times \vec{E} \right] \end{aligned} \quad (1)$$

where

$$\kappa = \frac{\alpha}{4\pi^2}, \quad \alpha = \frac{e^2}{\hbar c} \text{ being the fine structure constant,} \quad (2)$$

and  $\theta$  is the axion field. In the long wavelength limit where Rayleigh scattering is treated in the electro-magneto statics limit of (1), and in the absence of free sources, we have the simpler field equations reduced to the following:

$$\begin{aligned} \vec{\nabla} \cdot (\vec{D} + 4\pi\kappa\theta\vec{B}) &= 0 \\ \vec{\nabla} \cdot \vec{B} &= 0 \\ \vec{\nabla} \times \vec{E} &= 0 \\ \vec{\nabla} \times (\vec{H} - 4\pi\kappa\theta\vec{E}) &= 0 \end{aligned} \quad (3)$$

We shall assume all media including the TI are linear, homogeneous and nonmagnetic in their dielectric properties with  $\vec{D} = \epsilon\vec{E}$  and  $\vec{H} = \vec{B}$ . Furthermore, the axion field  $\theta$  is taken to be constant inside the TI and zero outside. With all these, one can then introduce two scalar potentials  $\phi$  and  $\phi_B$  where:

$$\vec{E} = -\vec{\nabla}\phi, \quad \vec{B} = -\vec{\nabla}\phi_B, \quad (4)$$

and both satisfy Laplace equations:

$$\nabla^2\phi = 0 \quad \text{and} \quad \nabla^2\phi_B = 0. \quad (5)$$

This 2-potential approach is analogous to the 2-vector potential formulation in [17] for Mie scattering in axion electrodynamics, and is equivalent to the ‘‘complex field approach’’ for the solution of chiral media [22].

In a similar way as in deriving the boundary conditions for the fields from the Maxwell’s equations without source, (3) leads to the following four boundary conditions [23]:

$$\epsilon_2 E_{2n} - \epsilon_1 E_{1n} = 4\pi\kappa\theta B_n \quad (6)$$

$$B_{1n} = B_{2n} = B_n \quad (7)$$

$$E_{1t} = E_{2t} = E_t \quad (8)$$

$$B_{2t} - B_{1t} = -4\pi\kappa\theta E_t, \quad (9)$$

where the subscript ‘‘1’’ refers to the TI and ‘‘2’’ the outside medium.

Next, we apply the results in (5)-(9) to the nanoshell as depicted in Fig. 1 where ‘‘1’’ is the TI core, ‘‘2’’ the plasmonic nanoshell, and ‘‘3’’ is simply vacuum. To study the TME on the coupled plasmonic resonances, it suffices to study merely the singular behavior of the electric polarizability of the system. Hence we assume the system being placed in a previously uniform electric field  $E_0$  along the  $\hat{z}$  direction and try to calculate the induced dipole polarizability of it. Thus the general solutions of (5) for each of the three regions can be expressed in standard

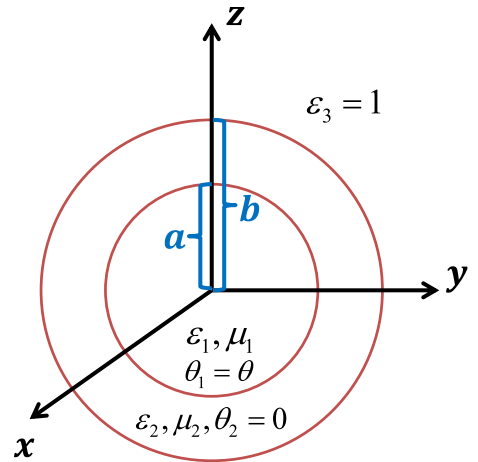


Fig. 1. Geometry of the plasmonic shell with a TI core.

form as:

$$\begin{aligned} \phi_1 &= \sum_{\ell} \alpha_{\ell} r^{\ell} P_{\ell}(\cos\theta) \\ \phi_{1B} &= \sum_{\ell} s_{\ell} r^{\ell} P_{\ell}(\cos\theta) \\ \phi_2 &= \sum_{\ell} (\alpha'_{\ell} r^{\ell} + \beta'_{\ell} r^{-\ell-1}) P_{\ell}(\cos\theta) \\ \phi_{2B} &= \sum_{\ell} (s'_{\ell} r^{\ell} + t'_{\ell} r^{-\ell-1}) P_{\ell}(\cos\theta) \\ \phi_3 &= -E_0 r P_1(\cos\theta) + \sum_{\ell} \beta''_{\ell} r^{-\ell-1} P_{\ell}(\cos\theta) \\ \phi_{3B} &= \sum_{\ell} t''_{\ell} r^{-\ell-1} P_{\ell}(\cos\theta) \end{aligned} \quad (10)$$

To solve the boundary value problem, we first note that, due to the asymptotic uniform field  $E_0\hat{z}$ , it is not difficult to anticipate that only dipole terms with  $\ell = 1$  will survive in all the potentials in (10) once the boundary conditions (6)–(9) are imposed. Furthermore, the two conditions for tangential components in (8) and (9) simply lead to analogous conditions for the corresponding potentials as in the ordinary electrostatics case. Thus with the application of (6)–(9) at the outer boundary of the shell ( $r = b$ ), we obtain (with only  $\ell = 1$  terms and  $\theta_2 = 0$ ):

$$\begin{aligned} 2\beta'_{\ell}\epsilon_2 b^{-3} - \epsilon_2\alpha'_{\ell} - 2\beta''_{\ell} b^{-3} &= E_0 \\ 2b^{-3}t''_{\ell} + S'_{\ell} - 2b^{-3}t'_{\ell} &= 0 \\ b^{-3}\beta''_{\ell} - \alpha'_{\ell} - b^{-3}\beta'_{\ell} &= E_0 \\ b^{-3}t''_{\ell} - S'_{\ell} - b^{-3}t'_{\ell} &= 0 \end{aligned} \quad (11)$$

Similarly, application of (6)–(9) at the inner boundary of the shell ( $r = a$ ), we obtain (with only  $\ell = 1$  terms and  $\theta_1 = \theta = (2N + 1)\pi$ ):

$$\begin{aligned} \epsilon_2\alpha'_{\ell} - 2\epsilon_2 a^{-3}\beta'_{\ell} - \epsilon_1\alpha_{\ell} - 4\pi\kappa\theta S_1 &= 0 \\ S_1 + 2a^{-3}t'_{\ell} - S'_{\ell} &= 0 \\ \alpha'_{\ell} + a^{-3}\beta'_{\ell} - \alpha_{\ell} &= 0 \\ S'_{\ell} + a^{-3}t'_{\ell} - S_1 + 4\pi\kappa\theta\alpha_1 &= 0 \end{aligned} \quad (12)$$

where we have adopted odd multiple values of  $\pi$  for the axion angle for TI with broken time reversal symmetry following previous works in the literature [24,25]. The combined linear system with (11) and (12) contains eight unknowns, namely,  $\alpha_{\ell}$ ,  $\alpha'_{\ell}$ ,  $\beta'_{\ell}$ ,  $\beta''_{\ell}$ ,  $s_1$ ,  $s'_{\ell}$ ,  $t'_{\ell}$  and  $t''_{\ell}$ . For the polarizability of the nanoshell with the TI core, we have to solve the system for  $\beta''_{\ell}$ . Straightforward algebra finally leads to the following result:

$$\beta''_{\ell} = \frac{[2(\epsilon_2 - 1)b^3 - (2\epsilon_2 + 1)a^3]\epsilon_2 + [(2\epsilon_2 + 1)a^3 + (\epsilon_2 - 1)b^3][\epsilon_1 + 2(4\pi\kappa\theta)^2/3]}{[2(\epsilon_2 + 2)b^3 - 2(\epsilon_2 - 1)a^3]\epsilon_2 + [2(\epsilon_2 - 1)a^3 + (\epsilon_2 + 2)b^3][\epsilon_1 + 2(4\pi\kappa\theta)^2/3]} b^3 E_0 \quad (13)$$

Hence the electric dipole polarizability of the nanoshell with a TI core is obtained as:

$$\alpha_{ee} = \frac{[2(\varepsilon_2 - 1)b^3 - (2\varepsilon_2 + 1)a^3]\varepsilon_2 + [(2\varepsilon_2 + 1)a^3 + (\varepsilon_2 - 1)b^3][\varepsilon_1 + 2(4\pi\kappa\theta)^2/3]}{[2(\varepsilon_2 + 2)b^3 - 2(\varepsilon_2 - 1)a^3]\varepsilon_2 + [2(\varepsilon_2 - 1)a^3 + (\varepsilon_2 + 2)b^3][\varepsilon_1 + 2(4\pi\kappa\theta)^2/3]} b^3. \quad (14)$$

In the limit of a nanoshell with an ordinary dielectric core,  $\theta = 0$  and (14) reduces to the well-known result in the literature [26]. For the case of a bare TI sphere of radius  $a$ , one simply sets  $\varepsilon_2 = 1$  and (14) reduces to [27]:

$$\alpha_{SP} = \frac{\varepsilon_1 - 1 + 2(4\pi\kappa\theta)^2/3}{\varepsilon_1 + 2 + 2(4\pi\kappa\theta)^2/3} a^3. \quad (15)$$

In the following, we shall use the result in (14) to study the TME effects on the coupled plasmon modes of a plasmonic nanoshell.

## Results and discussion

To study numerically the TME on the coupled plasmon resonances, we shall assume all materials to be nonmagnetic and adopt a single-mode model for the dielectric function of the topological insulator core given by [24,25]:

$$\varepsilon_1 = 1 - \frac{\omega_c^2}{\omega^2 - \omega_R^2} \quad (16)$$

and for the plasmonic shell, we shall assume the validity of the Drude model applied to the following two cases [28]:

Case (i): A silver shell of free electrons with  $\varepsilon_2$  given by [26]:

$$\varepsilon_2 = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} \quad (17)$$

where we have the plasmon frequency at the UV range with  $\omega_p = 1.36 \times 10^{16} \text{Hz}$ , and the damping constant given by  $\Gamma = \Gamma_{\text{Bulk}} + \left(\frac{Av_F}{b-a}\right)$ , with  $\Gamma_{\text{Bulk}} = 2.56 \times 10^{13} \text{Hz}$ ,  $v_F = 1.39 \times 10^{15} \text{nm/s}$ , and set  $A = 1$ .

Case (ii): A doped silicon shell with  $\varepsilon_2$  given by [29]:

$$\varepsilon_2 = 11.7 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} \quad (18)$$

with the THz plasmon frequency given by  $\omega_p = 2.1 \times 10^{13} \text{Hz}$  and  $\Gamma = 5.3 \times 10^{12} \text{Hz}$ .

In addition, the universal TME parameter has the value given by [24,25]:

$$4\pi\kappa\theta = \alpha\theta \sim \frac{\theta}{137} = \frac{(2N+1)\pi}{137} \quad (19)$$

with  $\alpha$  the fine structure constant and  $N$  an integer for a TI with broken time-reversal symmetry.

Fig. 2 shows the results for the imaginary part of  $\alpha_{ee}$  in Eq. (14) as a function of frequency for a silver nanoshell of dimensions  $a = 8 \text{ nm}$  and  $b = 10 \text{ nm}$ , while the parameters in Eq. (16) for the TI are taken from Ref. [24] with  $\omega_R = 56 \text{ cm}^{-1}$  ( $\sim 1.6 \text{ THz}$ ) and  $\varepsilon_1(0) = 4$ , corresponds to  $\omega_c = \sqrt{3}\omega_R$ . The results are shown for several values of the axion angle according to  $\theta = (2N+1)\pi$ . It is of interest to observe that while the bonding (low-frequency) and anti-bonding modes of the metallic nanoshell respond quite differently in their absorption strengths with a decrease (increase) for the bonding (anti-bonding) mode strength as the TME effect increases; both modes experience increasing observable red-shifts arising from the TME effects. In addition, the bonding modes are seen to be more sensitive to these TME effects as shown in Fig. 3 where the red-shifts are plotted for each of the two coupled plasmon modes. This could be due to the fact that the bonding modes are being closer to the characteristic frequency of the TI which is in the THz regime. In addition, the effects on the bonding modes obtained from our application of the AED are more justified since AED as an effective theory for TI is in general more accurate in the limit of low frequencies [11]. Also shown in the Figure in broken lines are the results predicted from using an idealized Drude model for the metal (see

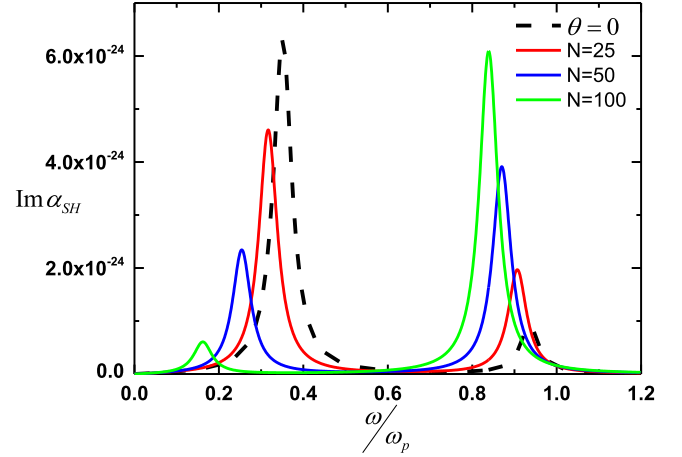


Fig. 2. Imaginary part of the dipole electro-electric polarizability of the shell ( $\alpha_{SH}$ ) as a function of frequency. Note that the case of a nontopological core is shown in broken line.

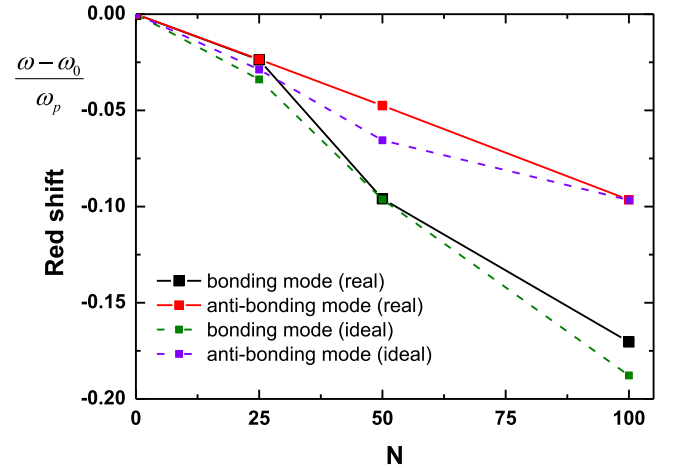


Fig. 3. Red-shift (normalized to the plasmon frequency) in plasmon resonances for a silver nanoshell with a TI core. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Appendix A) with the TI dielectric function set to unity. It is seen that the effects due to the damping of the metal and the dispersion of the TI are minimum in this case.

It is of interest to trace the origin of these universal red-shifts in the plasmonic frequencies due to the TME effects. In fact, in a previous investigation of the surface plasmon at a planar metal-TI interface, Karch [28] has also obtained similar red-shifts which depend on the square of the axion angle. Here our focus is on the *coupled* plasmon modes of a spherical nanoshell. Moreover, if one goes through the derivation of Karch's work, it is not difficult to conclude that such red-shifts are originated from the *different signs* in the axion term which appears in the *effective* constitution relations for the displacement and magnetic fields [11]:

$\vec{D} = \varepsilon\vec{E} + (\alpha\theta/\pi)\vec{B}$  and , respectively (note that the  $P_3$  field in [11] is related to our axion field via:  $P_3 = -\frac{\theta}{2\pi}$ ). This difference in signs in  $\vec{D}$  and  $\vec{H}$  can further be associated with the standard feature of duality transformation in electromagnetism, and is consistent with the generalized dual-symmetric theory of AED for TI as formulated by Karch [29]. It will be of interest to further investigate the physical origin of this universal red shifts.

In order to have the shell plasmonic frequency closer to the THz range, we next consider a doped silicon shell as reported in [30] which

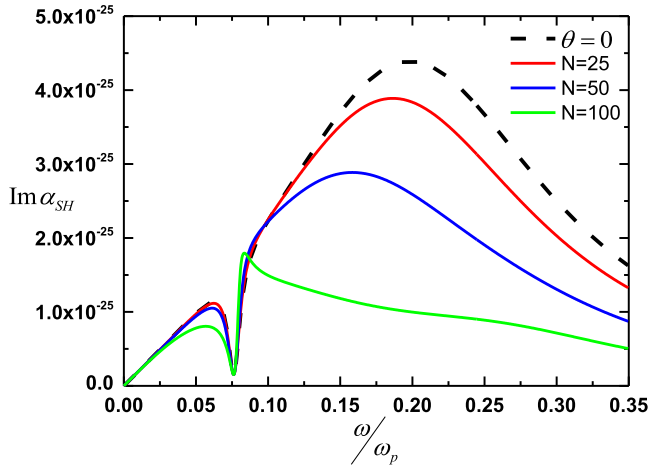


Fig. 4. Same as in Fig. 2, but for a doped silicon shell.

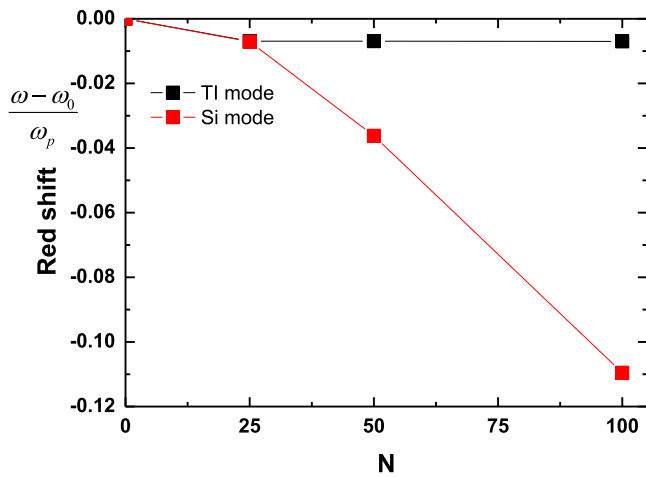


Fig. 5. Same as in Fig. 3, but for a doped silicon shell.

can be described by a Drude model as given in Eq. (18). The dimensions of the shell and the TI are the same as those in Fig. 2. In this case, we see

## Appendix A

When the damping in the Drude model  $\varepsilon_2$  in Eq. (17) and the dispersion of that for the TI in Eq. (16) are neglected, the resonance frequencies can be solved analytically by setting the denominator of Eq. (14) to zero with  $\varepsilon_1 = 1$  and  $\varepsilon_2 = 1 - \omega_p^2/\omega^2$ . The resonance frequencies for the coupled bonding and antibonding modes of the TI-filled metallic nanoshell can finally be obtained in the following form:

$$\omega_{\pm} = \frac{\omega_p}{\sqrt{3}} \sqrt{\frac{27 + (2 + 4x^3)\beta \pm [81(1 + 8x^3) - 36(1 - 10x^3)\beta + 4(1 + 2x^3)^2\beta^2]^{1/2}}{2(9 + 2\beta)}}. \quad (\text{A1})$$

In (A1),  $x \equiv a/b$ ,  $\beta \equiv (4\pi\kappa\theta)^2$  and the plus (minus) sign inside the square root refers to the antibonding (bonding) modes, respectively. The result in (A1) reduces to the well-known results in the limit when  $\beta$  and/or  $x$  vanishes. It can be shown in either case, (A1) with  $\beta \neq 0$  leads to red-shifts in resonance frequencies as shown in Fig. 3 by the broken lines.

## Appendix B

Here we give the results for the other polarizability functions for the TI-filled nanoshell which will be useful for the calculation of the cross sections for Rayleigh scattering from such a system [18,19]. These can be obtained by introducing also a uniform magnetic field into the present formulation, and in a similar and slightly more tedious analysis, we obtain the following results for the electro-magnetic, magneto-electric, and magneto-magneto polarizabilities of the TI-filled nanoshell, with the electro-electric polarizability as already given in Eq. (14):

$$\alpha_{em} = \frac{12\varepsilon_2 a^2 \pi \kappa \theta}{[2(\varepsilon_2 + 2)b^3 - 2(\varepsilon_2 - 1)a^3] \varepsilon_2 + [2(\varepsilon_2 - 1)a^3 + (\varepsilon_2 + 2)b^3][\varepsilon_1 + 2(4\pi\kappa\theta)^2/3]} b^3, \quad (\text{B1})$$

from Fig. 4 that the shell plasmonic modes do not split due to the relatively large damping of the shell material. The low frequency peak emerged has its origin actually from the absorption resonance of the TI. Moreover, the unsplit plasmon mode still undergoes the universal red-shift induced by the TME effect, whereas the TI absorption peak is essentially unaffected by such shifts. The red-shift for the plasmon resonance is again shown in Fig. 5 which is seen to have an observable value comparable to the case of the metallic shell.

## Conclusion

In this work, we have demonstrated that surface plasmonic modes from a metallic or doped semiconductor nanoshell can be utilized as a probe for the TME if the core of such a shell is replaced by a TI. The universal red-shift which depends only on the axion angle and the fine structure constant is seen to be more sensitive for the low frequency bonding mode of the metal shell, and is speculated to be a manifestation of the duality symmetry as generalized for axion electrodynamics [29]. Moreover, recent progress has demonstrated that it is now possible to fabricate TI with plasmonic frequencies in the optical range, bringing such frequencies closer to those of the metallic plasmons [31].

As far as we are aware, experimental observation of such TME-induced universal red-shifts in plasmonic frequencies has not yet been reported in the literature. One possible way is to perform optical scattering experiments from such TI-filled nanoshells. To account for such a process, one would need to calculate also the electro-magnetic, magneto-electric and magneto-magnetic polarizabilities [17,19,22]. Generalization of the previous calculations for a bare TI sphere to a shell geometry is straightforward using the two-potential formalism of the present work and the results are given in Appendix B.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgment

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$$\alpha_{me} = \frac{12\varepsilon_2 b^3 \pi \kappa \theta}{[2(\varepsilon_2 + 2)b^3 - 2(\varepsilon_2 - 1)a^3]\varepsilon_2 + [2(\varepsilon_2 - 1)a^3 + (\varepsilon_2 + 2)b^3][\varepsilon_1 + 2(4\pi\kappa\theta)^2/3]} a^3 = \alpha_{em}, \quad (\text{B2})$$

$$\alpha_{mm} = -\frac{[2(\varepsilon_2 - 1)a^3 + (\varepsilon_2 + 2)b^3](4\pi\kappa\theta)^2/3}{[2(\varepsilon_2 + 2)b^3 - 2(\varepsilon_2 - 1)a^3]\varepsilon_2 + [2(\varepsilon_2 - 1)a^3 + (\varepsilon_2 + 2)b^3][\varepsilon_1 + 2(4\pi\kappa\theta)^2/3]} a^3, \quad (\text{B3})$$

Note that the symmetry between  $\alpha_{em}$  and  $\alpha_{me}$  is different than that for the case of a chiral sphere (see Lindell et al in Ref. [22]) since the dual symmetry in AED is different from that for conventional electrodynamics [29].

### Appendix C. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.rinp.2019.102744>.

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