# Surface Plasmon Resonance (Spr) Sensor Simulation with Kretschmann Configuration Based on Chitosan-Graphene Oxide Nanocomposite for Lead Metal Ion Detection

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**ABSTRACT**

The presence of metal ions in river water can have a negative impact on humans when consumed. The detection of metal ions is crucial, and one of the methods is the surface plasmon resonance (SPR) phenomenon using SPR sensor system. SPR biosensor system can be used for metal ion deteection, offering advantages such of affordability and rapid detection. This research aims to investigate the influence of adding chitosan-graphene oxide (CS-GO) nanocomposites to SPR sensors as lead ion detectors. The CS-GO nanocomposites is used as a detector with variations in volume fraction and thickness to find the optimum values. The varied volume fractions are 18 nm, 20 nm and 25 nm. The involvement of the CS_GO nanocomposites is demonstrated through the reflectance curve, showing the SPR angle and reflectance values. The Winspall simulator is used to display the reflectance curve. The SPR system utulized a He-Ne laser with a wavelength of 632,8 nm, a BK7 half cylinder prism, and a thin film of silver metal layer. The SPR sensor configuration used is the Kretschmann configuration. The layer system models include BK7 prism/Ag thin film/chitosan/lead ion, and BK7/Ag thin film/CS-GO/lead ion. The nanocomposite CS-GO's pernittivity is calculated using the Maxwell Garnett effective medium theory. Result show that the refractive index and permittivity values of the CS-GO nanocomposite increase with the increasing volume fraction. The nanocomposite with a volume fraction of 0,099 produce a permittivity of 2,4337 and a refractive index of 1,5600.Larger permittivity values and thicknesses of the nanocomposite also lead to a shift in the SPR angle in the reflectance curve. The most significant SPR angle shift is observed with the use of CS-GO nanocomposite with a volume fraction of 0,099 and a thickness of 25 nm.



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## 1. INTRODUCTION

The increasing industrial activities have lead to environmental issues such as river water pollution. Industries such as painting, mining, and metal smelting often use heavy metal ions like lead and copper [1][2]. Direct industrial waste disposal inti rivers result in elevated levels of heavy metal ions in river water, posing a threat to human health [3][4]. High concentrations of heavy metal ions in river water have negative effect on aquatic organism as well as humans [5]. Heavy metals that enter the human body can be hazardous to health, as they can inhibit enzume function, disrupt bodily metabolism, and lead to conditions like cancer and genetic mutations [6][7]. Even at very low concentrations, lead ions can damage the brain, central nervous system, and kidney in both adults and children [8][9].

The increase in heavy metal levels in river water is a significant issue, as rivers serve as a vital source of drinking water for humans. Therefore, the detection of heavy metals in river water before its consumption is crucial. Several methods can be employed to detect heavy metal ions in river water, such as atomic absoption spectrometry, inductively coupled plasma mass spectrometry, impregnated strip and X ray fluorecence spectrometry [10][11]. While these methods are commonly used, they come with certain drawbacks such as expensive equipment costs, complex sample processing, ang lengthy measurement times. Apart from these methods, there's another approach that is more affordable, offers quick measurement times, and boasts high selectivity, that is the surface plasmon resonance (SPR) sensor [12][13].

SPR is a physical process that occurs when polarized light propagates through a prism and penetrates a thin metal layer under conditions of total internal reflection. Under the circumstance, a wave is generated that can propagate into a dielectric medium. At a specific angle of incidence, when the light wave vector matches the wave vector of the surface plasmon wave, reflectance experiences a significant decrease [14] The fundamental principle of SPR biosensors as sensors relates to the oscillations of free electrons at the boundary between metal and dielectric medium, known as surface plasmon waves [15][16]. Surface plasmon (SP) waves are electromagnetic waves that propagate at the interface between two media with different refractive indices and can be induced using the attenuated total reflection (ATR) method [17]. These two media consist of a metal layer on the prism surface and a thin biomolecular layer (dielectric medium) on the metal surface that serves to detect analytes [18]. The operation principle of SPR begins when evanescent and surface plasmon waves couple or resonate. This resonance condition is expressed in equation 1 [19][20].

$$\frac{\omega_0}{c} n_p \sin\theta_{SPR} = \frac{\omega_0}{c}\left(\frac{\varepsilon_m n_d^{\,2}}{\varepsilon_m + n_d^{\,2}}\right)^{1/2} \tag{1}$$

The variables on the left-hand side are the propagation constant of the light beam at the resonance angle that passes through the prism with a refractive index . Meanwhile, the variables on the right-hand side are the propagation constant with being the real component of the metal's permittivity and being the refractive index of the dielectric material or sensing material. ω0 and c respectively represent the frequency of light and the speed of light in a vacuum [8]. Physically, SPR can be observed through a significant decrease in reflectance at a specific angle of incidence accompanied by energy loss. In SPR sensors, thin metal layers like thin silver layers are commonly used to excite surface plasmon waves. However, the use of thin silver layers still has disadvantages, such as susceptibility to oxidation and sulfidation. Thus, other protective materials need to be added, such as the chitosan-graphene oxide (CS-GO) nanocomposite [21]. Lookman et al. detected lead ions using an SPR sensor with both thin silver layers and CS-GO nanocomposites. The study was conducted experimentally using lead ion samples ranging from 0,03 to 5 ppm with a constant volume fraction. The research successfully detected lead ions using SPR, producing larger SPR angles for higher lead ion concentrations [22].

. In this study, SPR simulation for lead ion detection will be performed using the Winspall software. The use of Winspall requires data such as the refractive index and material thickness. Some materials to be used, such as BK7 prism, thin silver layer, and lead ions, already have known refractive index values from various references. However, the refractive index value of the CS-GO nanocomposite is unknown, necessitating the calculation of its refractive index value. The volume fraction of the CS-GO nanocomposite will be varied, while the concentration of the detected lead ions remains constant.

## 2. METHODS

In this study, the first step is to determine the SPR system model involving multiple materials using the Kretschmann configuration [19]. The SPR system is modeled with two configurations, involving four layers: BK7 prism, Ag thin film, CS nanocomposite, and the analyte, as depicted in Figure 1(a), and BK7 prism, Ag thin film, CS-GO nanocomposite, and the analyte, as shown in Figure 1(b).

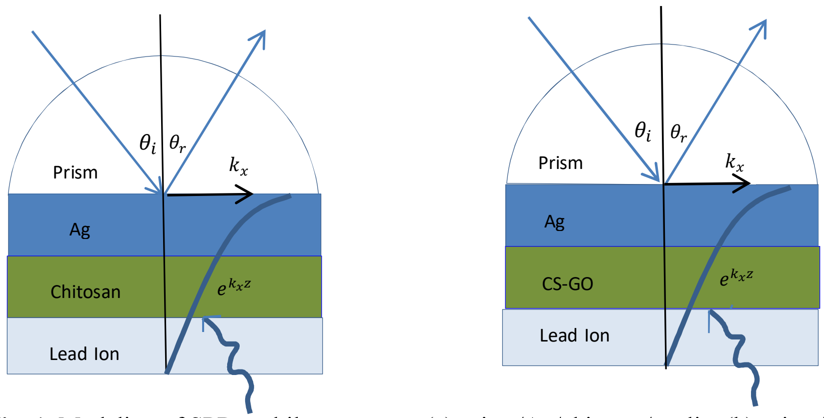


**Fig. 1.** Modeling of SPR multilayer system (a) prism/Ag/chitosan/analite (b) prism/Ag/nanocomposite CS-GO/analite

The wavelength used in the SPR system is 632.8 nm (visible light). The optimal thickness of the thin silver layer that produces a sharp spectrum is 40 nm [23]. The simulation parameters used are indicated in the following Table 1:

**Table 1.** Refractive index values and Thicknesses of each layer

| Material | Refractive index | Thickness |
|---|---|---|
| **BK7 prism [23]** | 1,51 | - |
| **Ag thin film [23]** | 0.13455+3,98651i | 40 nm |
| **Chitosan [24]** | 1,52970 | 18 nm |
| **GO [25]** | 3,83 | |
| **CS-GO nanocomposite** | Variated result from determination | 25 nm |
| **Lead Ion 0,5 ppm [26]** | 1,3317+0,0002i | - |

The use of thin silver layers in SPR biosensors has a drawback, which is their susceptibility to oxidation. Therefore, it's necessary to add another material as a protective layer over the thin silver layer. In this context, chitosan and the CS-GO nanocomposite act as protective layers over the thin silver layer while also serving as the sensing layer to detect analytes. In this research, the detected analyte is lead ions at a concentration of 0.5 ppm, which has a complex refractive index of 1.3317+0.0002i [26].

The next step involves calculating the effective permittivity value of the CS-GO nanocomposite, which can be done using the Maxwell-Garnett equation [27]. This equation is an algebraic formula that expresses the effective permittivity of a composite material as a function of the permittivity of its constituents, volume fraction, and other parameters that define the microscopic structural characteristics of the composite material. This composite can consist of homogeneous inclusion materials embedded in another homogeneous material, making its permittivity function continuous. Maxwell-Garnett models a composite or mixed material as a heterogeneous material composed of inclusion and matrix materials. In this case, graphene oxide acts as the inclusion material, while chitosan serves as the matrix material. The Maxwell-Garnett equation is expressed by Equation 2 [27].

$$\varepsilon_{eff} = \varepsilon_m \frac{\varepsilon_m + [v_p(1-f) + f](\varepsilon_i - \varepsilon_m)}{\varepsilon_m + v_p(1-f)(\varepsilon_i - \varepsilon_m)} \tag{2}$$

where , and are the effective permittivity of the CS-GO nanocomposite, matrix permittivity (chitosan), and inclusion permittivity (graphene oxide), respectively.represents the volume fraction of the inclusion, and is the depolarization factor of the ellipsoid, which is typically set to 1. The inclusion volume fraction is varied as 0.062, 0.082, and 0.099. Furthermore, the complex refractive index values for each material involved in the SPR system can be obtained using Equation 3.

$$n = \sqrt{\varepsilon} \quad (3)$$

where and is complex refractive index and permittivity

The SPR simulation is conducted after calculating the effective permittivity for each variation of the composite volume fraction. Complex permittivity values for BK7 prism, silver, and lead ion are known. All these complex permittivity parameters along with layer thicknesses are input into Winspall 3.02 software [28][29]. The SPR phenomenon is depicted by the Attenuated Total Reflection (ATR) curve, which displays a significant decrease in reflectance at a specific angle of incidence. This minimum angle of reflectance is referred to as the SPR angle. The sharp dip in the ATR curve (SPR angle) signifies the energy absorption by the surface plasmon (excited electrons), leading to a reduction in the reflected light energy and consequently creating a minimum reflectance point [30]. The shift of the SPR angle to the right in the ATR curve indicates that the modeled sensor system is capable of detecting the presence of the analyte, in this case, lead ions.

## 3. RESULTS AND DISCUSSION

The values of the permittivity and refractive index of the CS-GO nanocomposite for various inclusion volume fraction variations obtained through calculations using Equation 1 can be seen in Table 2 as follows:

**Table 2.** The values of refractive index and permittivity of CS-GO nanocomposite

| No | Volume Fraction variation | Permittivity | Refractive index |
|---|---|---|---|
| **1** | 0,062 | 2,3978 | 1,5485 |
| **2** | 0,082 | 2,4171 | 1,5547 |
| **3** | 0,099 | 2,4337 | 1,5600 |

Table 2 demonstrates that the refractive index and permittivity values of the CS-GO nanocomposite are influenced by the inclusion volume fraction. As the inclusion volume fraction increases, the composite's refractive index and permittivity values also increase. This indicates that adding more GO volume to the composite results in higher refractive index and permittivity values for the composite.

The SPR phenomenon can be observed through the ATR curve. The SPR system for lead ion detection without involving the composite is shown in Figure 2(a), indicating an SPR angle of 45.7021°. Meanwhile, the SPR system involving the CS-GO composite is depicted in Figures 2(b)-2(c) for inclusion volume fraction variations of 1 to 3, revealing a shift in the SPR angle: 45.7861°, 45.8100°, and 45.8297°, respectively.

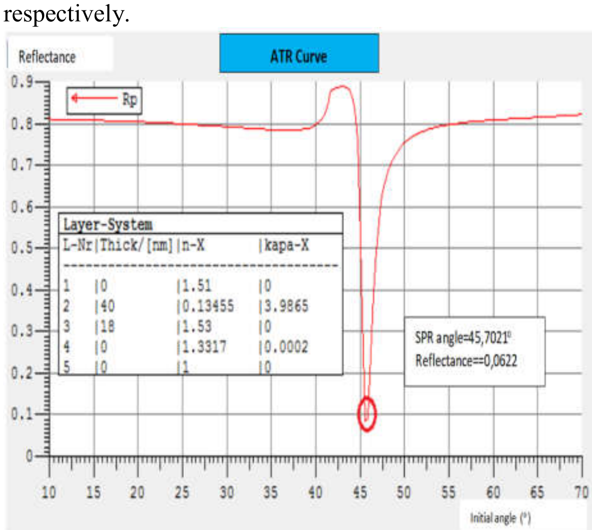


**Fig. 2.(a)** Kurva ATR Curve for the biosensor system prism/BK7/Ag thin Film /kitosan/ion timbal

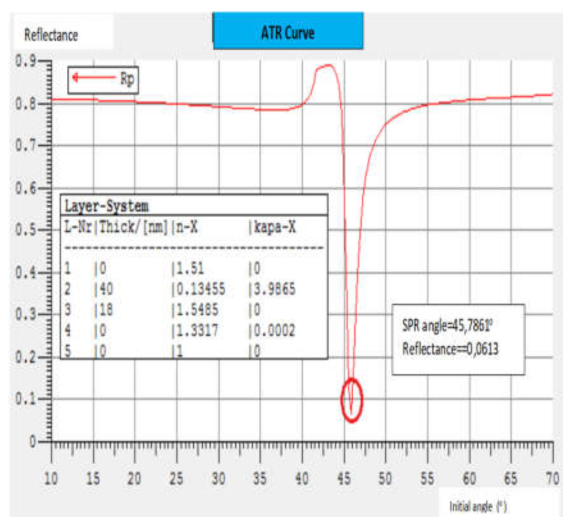


**Fig. 2.(b)** ATR curve for the biosensor system :BK7 prism/Ag thin film/CS-GO nanocomposite with a volume fraction of 1/ion lead

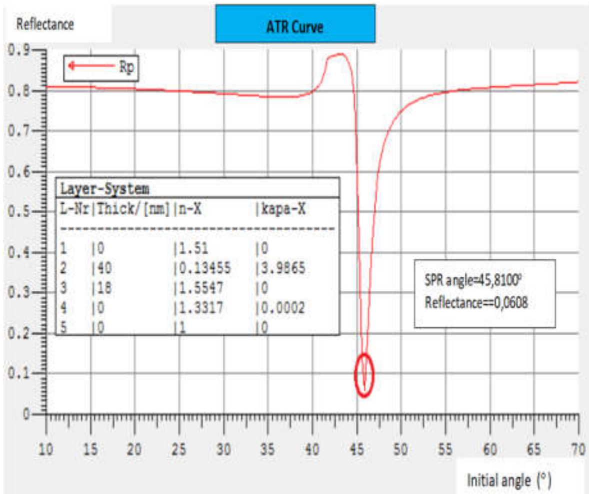


**Fig. 2.(c)** ATR curve for the biosensor system :BK7 prism/Ag thin film/CS-GO nanocomposite with a volume fraction of 2/ion lead

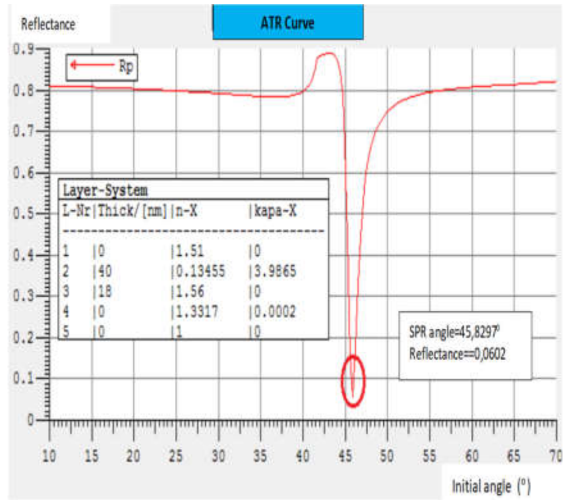


**Fig. 2.(d)** ATR curve in the biosensor system: BK7 prism/Ag thin film/CS-GO nanocomposite with a volume fraction of 3/lead ion

On the other hand, the SPR system for lead ion detection using variations in the thickness of the CS-GO composite is shown in Figures 3(a) and 3(b), where the SPR angles are 46.2033° and, subsequently, shifting to the right at an angle of 47.2466°.

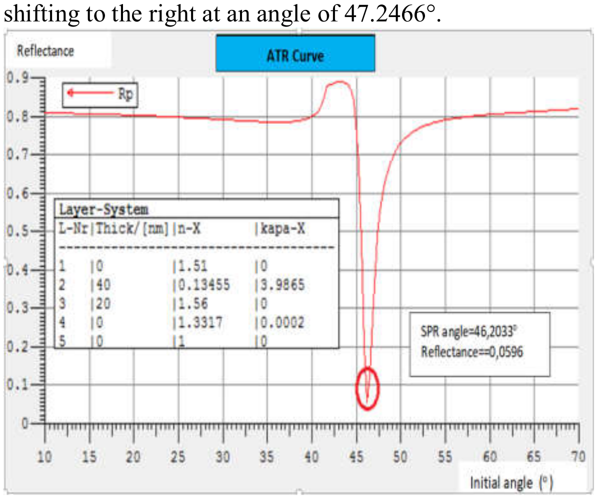


**Fig. 3.(a)** The Attenuated Total Reflection (ATR) curve for the sensor system with the configuration BK7 prism/Ag thin film/CS-GO composite with a thickness of 20 nm/lead ion

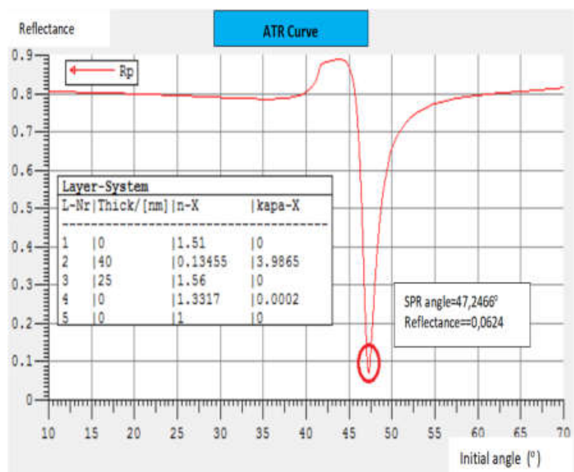


**Fig. 3.(b)** The Attenuated Total Reflection (ATR) curve for the sensor system with the configuration BK7 prism/Ag thin film/CS-GO nanocomposite with a thickness of 25 nm/lead ion

The influence of adding CS-GO nanocomposite to the SPR sensor for lead ion detection with variations in the composite's volume fraction can be observed through the shift in the summarized ATR curve presented in Figure 4. In this volume fraction variation, the largest SPR angle value is exhibited by the volume fraction of 0.099. This implies that as more GO is added to the composite, it results in a larger SPR angle for lead and copper ion detection. Further observation is conducted with the sensor system using the configuration BK7 prism/thin silver layer/CS-GO nanocomposite/lead ion with a constant volume fraction of 0.099, and variations in the nanocomposite's thickness of 20 nm and 25 nm, as shown in Figure 5. This figure reveals that as the composite layer thickness increases to 35 nm, the SPR angle shifts further to the right compared to a thickness of 20 nm. This doesn't necessarily indicate that the thicker the layer, the more the

SPR angle will shift to the right. Instead, there exists an optimum thickness value that reflects the best performance of the constructed SPR system.

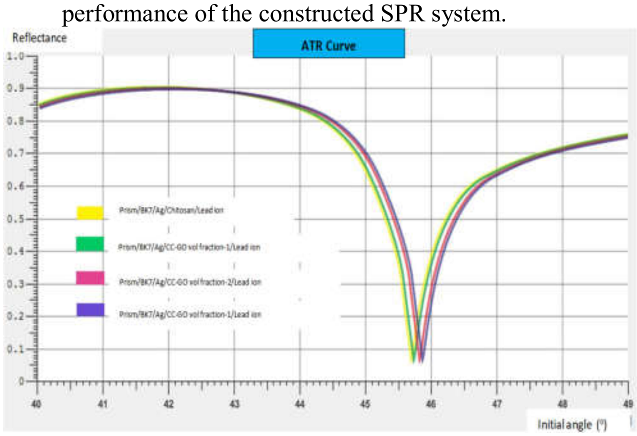


**Fig. 4.** The ATR curves in the biosensor system before and after the addition of CS-GO nanocomposite with varying volume fractions for lead ion analyte

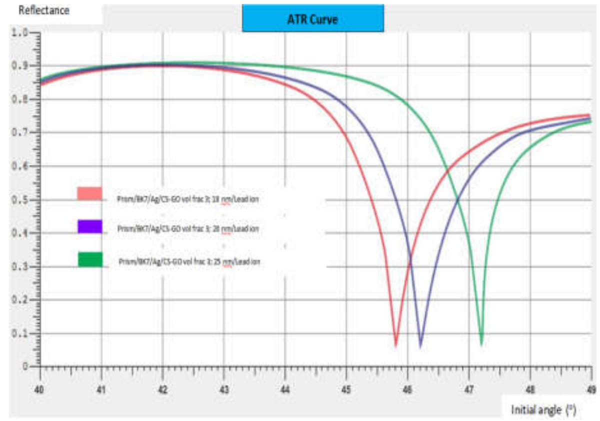


**Fig. 5.** The ATR curves in the biosensor system with variations in the thickness of the CS-GO nanocomposite for lead ion analyte

Based on the dispersion relation equation where the resonance or coupling between the evanescent wave and the surface plasmon wave occurs through Equation 1, the SPR angle is Depending on the values of complex refractive index parameters and the thickness of each layer involved in the modeled SPR system, the resonance or coupling between the evanescent wave and the surface plasmon wave occurs through Equation 1. The SPR angle is a result of the resonance between the evanescent wave and the surface plasmon wave, and its value depends on the refractive index of the prism used (BK7 prism), the refractive index of the thin metal layer (thin silver layer), and the refractive index of the dielectric material above the thin metal layer. If there are changes in the refractive index of any of these materials, the SPR angle will shift accordingly. It can be said that this SPR system is highly sensitive to changes in the refractive index of materials. This characteristic forms the basis of the application of SPR biosensors in detecting biomolecule concentrations. The rightward shift of the ATR curve indicates that there's a difference in the SPR angle values for the biosensor system before and after the addition of CS-GO nanocomposite. This is due to the difference in refractive index values between chitosan and the nanocomposite. This discrepancy in refractive index values alters the position of the SPR angle, which is a representation of the coupling between the evanescent wave and the surface plasmon wave. Additionally, the volume fraction value in the CS-GO nanocomposite affects its refractive index value, causing a shift in the ATR curve.Several journals have mentioned that the increase in the SPR angle before and after the addition of CS-GO nanocomposite is related to the surface roughness of both chitosan and CS-GO nanocomposite surfaces. Lokman et al. (2014) and Lokman et al. (2019) stated that the CS-GO nanocomposite has a higher surface roughness compared to chitosan. This increased surface roughness implies a higher surface-to-volume ratio, which is advantageous for binding lead ions. This characteristic is what causes the SPR system after the addition of CS-GO nanocomposite to yield a larger SPR angle compared to the system before the addition of CS-GO nanocomposite.

## 4. CONCLUSION

The values of permittivity and refractive index for the chitosan-graphene oxide nanocomposite have been obtained at volume fractions of 0.062, 0.082, and 0.099. These values increase as the volume fraction increases. The addition of the KS-GO nanocomposite in the SPR sensor system for metal ion detection can be observed through the shift in the ATR curve. The resulting SPR angle also increases as the volume fraction and thickness of the KS-GO nanocomposite are varied. The best performance is achieved when the SPR sensor exhibits a significant angle shift and low reflectance values. In this study, the KS-GO nanocomposite with a volume fraction of 0.082 and a thickness of 20 nm exhibited the lowest reflectance value, namely 0.0596. On the other hand, the largest SPR angle shift was obtained in the SPR system with the KS-GO nanocomposite at a volume fraction of 0.099 and a thickness of 25 nm. The resulting SPR angle was

47.2466°. To determine the optimal thickness value, further variations in thickness above 25 nm are necessary.

## DECLARATION

### Author Contribution

In this study, the first step is to determine the SPR system model involving multiple materials using the Kretschmann configuration.

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### Conflict of Interest

The authors declare no conflict of interest.

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